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On the Carrier Efficiency and Joint Toxic Action of Insecticidal Solvents. Insect Toxicological Studies on the Joint Toxic Action of Insecticides. IV.

AUTHOR(S):

SAKAI, Seiroku; ASUKA, Yoshimichi

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Résumé

This study deals with the population fluctuation of *Chilo suppressalis* from the capture record of light trap at Kusatsu (Shiga Pref.) for sixteen years, in relation to the condition of host plant and the meteorological factors.

First of all, the date at which half of the individuals is entrapped and the population in first generation are both correlated with these in the second generation of the preceding year. Such relations are not found between both generations in a year.

The population entrapped in the second generation is remarkably influenced by the meteorological conditions in July, the average of maximum air temperature in the second and last

decades, the average of air temperature of the second decade and the amount of precipitation of the third five-days. The population is fairly influenced by the population entrapped in the later period of the first generation, and the latter is determined by the 95% entrapped date.

The reproductive rate in the first generation, which is pretty correlated with the population entrapped in the second generation, is not correlated with the air temperatures in July, but remarkably influenced by the rainfall in the second and last decades, especially the amount of precipitation of the last decade in July. This rate correlates straightly with the height of rice plant for the fifth five-days in July, which protects the larvae from the heat caused from sun shine. The relation that the reproductive rate has remarkable minus correlation with the rate of stalk elongation in September is very complex to the host plant.

The reproductive rate in the second generation has considerable minus correlation with the rate in the first generation. The similar minus correlation with the parent population makes clear that the effect of population density is operative in the second generation where a little external environmental resistance is observed.

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20. 殺虫剤の溶剤の稀剤効果とその連合作用 殺虫剤の連合作用に関する昆虫毒物学的研究 第4報 酒井清六・飛鳥嘉道（八洲化学工業株式会社 研究所）31. 11. 10 受理

最も尊敬する春川忠吉先生の古稀の御祝いに本文を捧げる。

殺虫剤に使用される58種類の溶剤を夫々リンデン原末に1乃至2%添加した粉剤及び0.1%リンデン溶剤を使用して、リンデンに対する稀剤効果と連合作用とを研究した。供試昆虫は当研究所累代飼育のイエバエ及び野外より採集したエンマコオロギを使用した。粉剤試験には真空粉剤試験装置を使用し、0.1%リンデン溶剤試験にはエンマコオロギの左後脚を30秒間浸漬した。麻痺仰転した昆虫の致死率を時間の経過と共に観察した。大観して、芳香族炭化水素>脂肪族ケトン>ハロゲン化炭化水素は高い稀剤効果が認められた。然しグリコール、エステル及びアルコール類は低い稀剤効果であつた。稀剤効果と沸点との関係は凹状の曲線（エンマコオロギ）と山型曲線（イエバエ）とが認められた。蜜蠟及びイエバエのリポイドに対する溶解度の高い溶剤は高い稀剤効果が認められた。エンマコオロギに対する溶剤の表面張力と稀剤効果との関係はイエバエの時の関係より明瞭であつた。脂肪族一価アルコール、脂肪族ケトン及び芳香族炭化水素の溶剤の表面張力の実験では、稀剤効果は表面張力の上昇に伴い減少した。溶剤の粘度、極性、誘電恒数、双極子能率、分

子会合度と稀剤効果との関係を検討したが明瞭な結果は得られなかった。脂肪族一価アルコール群の安定したイエバエの致死率は炭素数 C_1 から C_6 の間で漸次上昇し、 C_6 に頂点を有し、その後 C_7 で下降した。これらの実験で得た結果は連合作用の分析を実施した。連合作用は一般に高い稀剤効果から低い稀剤効果のものが認められた。

Fundamental studies on the permeability of insect cuticle were investigated by numerous workers^{1,2,5-7,9-16,20,22-24,26,28-32}, but we have little analytical knowledge of insecticidal solvents concerned in cuticular permeability.

Fulton & Howard⁸ studied the effect of addition of oil on the toxicity to plant bugs of derris and other insecticides and pointed out that vegetable oils were said to be the more efficient insecticidal carriers. The oil solvents available as insecticidal solvents, whether vegetable or mineral, have higher lipo-solubility and destructive power of insect cuticle and are effective in increasing the permeability of insect cuticle.

Hurst¹¹ observed that the addition of paraffins and cycloparaffins to lindane were effective to the control of *Cimex*. He discovered that kerosene penetrates the cuticle of *Calliphora* larvae slowly and a polar solvent such as ethyl alcohol can penetrate rapidly. A mixture of apolar and polar solvent was effective in increasing the permeability of *Calliphora* larvae. Thus the effect of the kerosene has been interpreted as the destruction of lipid from its normal structural combination with protein in the lipoprotein mosaic of the *Calliphora* cuticle and the effect of the ethyl alcohol as increasing the permeability of hydrophilic parts. Considering the results obtained by Hurst^{13,14}, alcohol, ketones, fatty acids, amines, or phenols will aid the penetration of kerosene. When the speed of permeability to blowfly larvae was examined, Hurst found that butyl and amyl alcohol have carrier efficiency, whereas the higher alcohols are too surface active to be effective in this aspects.

Similar analysis of the effect of the solvent on the rate of penetration by the contact insecticides have been examined^{2,20,27,30-32}.

Webb & Green³⁰ found that the addition of 1% of certain solvents to an insecticidal powder containing 0.25% diphenylamine greatly reduced the time of death in *Melophagus ovinus*. Webb & Green pointed out that the ability of the

solvent to induce such an increase in rate of action was presumably due to an enhanced rate of penetration of insecticide through the sheep kid's cuticle and was termed the carrier efficiency of the solvent.

Recently, improved formulations to DDT and BHC emulsions such as Systron (composed of 15% lindane, and 85% solvents and detergents) are applied to the crop fields in Japan.

Concerning the improved formulations of chlorinated organic insecticides, Kaneko¹⁷ studied the action of fifteen solvents on the effectiveness of lindane emulsions and found the higher carrier efficiency of the mixture between methylnaphthalene and ketones solvents.

The present investigation was undertaken to demonstrate the joint action between organic solvents and lindane and to observe the effects of fifty eight solvents on the penetration of lindane powders independently to the work of Kaneko.

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Methods and techniques

The insect used were the adults of the common housefly, *Musca domestica vicina* Macq. and the adults of the oriental garden cricket, *Gryllus*

mitratus de Saussure. The houseflies used in this experiment were the progeny of the flies which have been reared for three years by inbreeding a small number of normal flies collected at Kōzu, Kanagawa.* The liquid of dried milk plus a small amount of sucrose was used as food for the adult and the mass culture medium for larval rearing consisted of 85% "Okarā", the residual product of bean-curd making, 4% dried yeast, 1% pepton, and 10% dried rice stem by weight. The insects were raised under a constant temperature of 30° and a relative humidity of 50%.

The crickets used in the experiment were collected at a pear orchard near Futago, Kawasaki. Cabbage leaves and dried fish powder were used as food.

The insecticide adopted was lindane. Fifty-eight insecticidal solvents were tested. The dust carrier mixture consisting of 6-parts clay and 4-parts talc (300 mesh) was applied to the dust formulations.

In the case of the fly test, about 50 houseflies at 3—4 days old after emergence were anaesthetized by carbon dioxide and placed on filter paper in a Petri dish provided with a covering of salan netting. The air-vacuum dust apparatus consisted of a glass jar, 15cm in inner diameter and 22cm high, with a glass plate covering its lower opening. The Petri dish containing about 50 flies was supported above the bottom of the glass jar. Air in the glass jar was produced by a rotary vacuum pump. 200 mg of the powder to be tested was placed in a glass tube with wire netting on its bottom. The glass tube was connected with the top tube of the jar. Then, 200 mg of powder was dusted into the apparatus under vacuum through the round hole of the top tube.

The flies were exposed to a cloud of dust for 30 seconds. After dusting, the insects were kept at a temperature of 28°. The number of paralysed flies were counted and the mortality was usually determined after 1, 2, 3, 4, 5, 18 and 24 hours.

In the case of the cricket test, the tarsus of the

cricket's hind leg was immersed into the test solution for 30 seconds. The test solution consisted of 0.1% lindane and 99.9% solvent. After the treatment, the insect was placed quickly on filter paper in the Petri dish and then the insect was at once transferred to the room surrounding. The time for paralysis after treatment was observed.

The adults of the housefly were crushed with a small glass stick, and then the dry matter extracted with ether for 24 hours, 22.6% yellowish lipoids per total fly by weight was obtained. The extract of lipoids was prepared for the experiments for the action of solvents on the housefly's lipoids. In order to measure the solubilities of the lipoids of the housefly to the solvents, one small drop (0.006g) of the fly lipid was placed in the Petri dish, 9 cm in diameter and 2 cm high. As soon as 5cc of the solvents for the experiments were poured slowly in the Petri dish, time in minute at which fly lipid was fully dissolved, was observed by the microscope.

The beeswax was also utilized for the experiments on the relationship between the carrier efficiency and the solubility of solvents to beeswax from the viewpoint of physiological significance. In order to measure the solubility of beeswax to solvents, one drop (0.02g) of liquid beeswax was introduced into 20cc Erlenmeyer flask, then the 5cc of solvents used for the experiments were poured in the Erlenmeyer flasks. The Erlenmeyer flask containing the solid beeswax and the solvent was at once shaken by an electric shaker at the rate of 167 shakes per minute. The time necessary for the solution of beeswax was observed.

The surface tensions of the solvents and the lindane-saturated solvents were measured by Du Nouey's surface tensions apparatus under a constant temperature of 30°.

The solubility of lindane in the solvents at a constant temperature of 30° was measured by weighing the lindane after the solvents evaporated and similar methods were used to measure the solubility of beeswax in solvents.

The effects of solvents of lindane on the mortality to the housefly

Using the housefly as a test insect, and applying

* We are indebted to Mr. K. Kojima, Towa Agricultural Chemicals Co. Ltd., for the supply of the original houseflies cultured for these tests.

to the cuticle insecticidal dust containing 0.1% lindane as the toxic agent, we observed that the additions of 1 and 2% of certain organic solvents greatly varied the time necessary for paralysis to occur.

Although we thought at first that the experiments for the effect of solvents on the permeability of lindane should be carried out the experiments for the liquid formulations, the analysis of the results obtained by the liquid formulation is so complex that water in the

emulsions tested is probably more limiting factor than the solvents in the emulsions.

In order to exclude the factor of water, we carried out the experiments on the dust formulations.

The results of the experiments on the relation between time and per cent paralysis in the fly for the solvents applied alone and with the lindane dusts added to the solvents are given in Tables 1 to 2.

As shown in Table 1, the addition of 2% organic

Table 1. The relation between time and per cent paralysis in the fly when 0.1% lindane dust added 1% and 2% solvents respectively against the adult of the housefly, *Musca domestica vicina* (the averaged values of 3 replicated trials). The number of fly tested: about 14,000. 28°C.

Solvent tested & Code no.	Solvent content % in w/w	Per cent paralysis in the fly at the elapsed time							Numerical values of the experimental curves		
		1hr.	2hr.	3hr.	4hr.	5hr.	18hr.	24hr.	N/2*	T* (minute)	N/2T*
K: Ketones (aliphatic, aromatic & cyclic)											
K-1 Acetone	1%			18.5		17.1	22.8	23.7	10.0	100.8	0.099
	2%			29.3		38.7	43.7	42.1	19.4	103.2	0.188
K-2 Acetophenone	1%	19.9	31.0	31.1	32.5	35.5	35.5	38.1	17.8	52.8	0.387
	2%	32.8	32.4	36.1	46.3	45.0	45.0	47.5	23.0	43.2	0.532
K-3 Acetylacetone	1%	29.0	20.7	21.2	23.7	24.1	27.9	27.6	12.0	31.2	0.385
	2%	25.7	26.6	25.8	34.0	30.5	34.3	35.5	15.3	26.4	0.580
K-4 Cyclohexanone	1%			48.8		35.6	36.0	37.0	18.1	50.4	0.361
	2%			49.8		44.5	42.1	42.7	21.4	57.6	0.372
K-5 Diacetone alcohol	1%	19.6	23.6	21.7	22.6	24.8	24.8	26.2	12.4	40.8	0.304
	2%	23.3	24.1	24.8	30.9	28.0	28.2	27.8	14.0	24.0	0.583
K-6 Di-ethyl ketone	1%	26.8	22.9	28.4	37.3	35.7	36.1	37.8	18.4	36.0	0.511
	2%	31.1	36.4	38.8	40.0	40.3	42.4	44.5	20.2	36.0	0.561
K-7 Di-iso-butyl ketone	1%	20.0	23.5	22.6	30.4	28.1	28.8	28.8	14.1	52.8	0.267
	2%	29.7	36.6	37.5	37.5	40.8	40.8	41.9	20.4	36.0	0.567
K-8 Methyleneethyl ketone	1%	29.4	48.8	44.3	45.7	43.9	41.9	43.8	21.9	48.0	0.452
	2%	53.0	63.0	60.8	64.9	65.2	68.9	65.5	32.5	19.2	1.693
K-9 Methyl-iso-butyl ketone	1%	11.9	10.5	8.6	11.3	14.2	14.2	15.5	7.1	36.0	0.197
	2%	32.6	45.1	46.9	62.1	64.2	70.2	72.8	34.0	69.6	0.489
p: Primary alcohols											
p-1 Methyl alcohol	1%			26.2		29.9	31.1	32.0	16.0	98.4	0.163
	2%			27.8		26.8	33.6	34.9	16.5	98.4	0.168
p-2 Ethyl alcohol	1%			24.9		28.5	28.5	28.5	14.3	76.8	0.186
	2%	25.0	30.0	30.0	32.5	35.5	35.0	35.0	17.5	33.6	0.521
p-3 n-Propyl alcohol	1%	19.4	43.3	38.8	39.7	39.0	39.3	37.9	19.4	67.2	0.289
	2%	29.3	41.4	41.2	46.0	45.4	46.9	48.5	23.0	48.0	0.479
p-4 iso-Propyl alcohol	1%			33.0		34.4	33.9	35.4	17.6	86.4	0.204
	2%			24.5		30.4	32.3	32.3	16.2	112.8	0.143
p-5 n-Butyl alcohol	1%	33.6	34.3	33.6		33.7	35.4	34.0	17.0	24.0	0.708
	2%	40.8	53.6	53.9		56.1	55.6	55.1	28.0	43.2	0.650
p-6 iso-Butyl alcohol	1%	39.7	45.7	47.3	45.4	44.5	50.2	48.3	23.0	28.8	0.799
	2%	45.6	63.4	61.9	69.8	72.1	74.8	75.5	36.5	43.2	0.845
p-7 iso-Amyl alcohol	1%	23.0	29.0	23.9	20.2	22.3	22.3	23.3	11.7	24.0	0.488
	2%	34.6	38.9	33.5	34.6	34.4	36.0	39.6	18.0	22.0	0.818

防 虫 科 学 第 22 卷—I

p-8 Benzyl alcohol	1%	26.1	35.6	32.1	32.4	29.6	29.8	30.1	14.8	33.6	0.440
	2%	35.6	39.9	40.9	41.6	44.7	47.0	46.8	23.0	31.2	0.737
p-9 Cyclohexanol	1%	34.8	50.5	49.2	47.0	40.7	44.1	41.5	20.6	26.4	0.780
	2%	55.8	69.8	66.2	68.9	68.6	72.3	66.9	34.5	24.0	1.438
h : Halogenated hydrocarbons											
h-1 Chloroform	1%	38.3	48.5	46.6		49.8	46.7	42.5	23.6	24.0	0.983
	2%	55.0	63.8	65.7		67.6	67.7	59.8	33.6	22.0	1.527
h-2 Carbon tetrachloride	1%	26.3	33.0	32.2		35.6	36.0	33.0	18.0	31.2	0.577
	2%	31.1	40.6	39.7		40.8	40.9	37.7	20.2	30.0	0.673
h-3 Ethylene dichloride	1%	33.7	42.4	41.7	43.5	41.9	40.8	41.1	21.0	40.8	0.515
	2%	46.6	54.8	53.4	56.0	56.5	61.5	58.7	29.0	33.6	0.863
h-4 Trichloroethylene	1%	28.6	39.3	38.0	38.2	37.4	40.5	40.5	20.2	43.2	0.468
	2%	39.6	50.8	49.4	48.2	47.9	58.8	58.3	26.0	33.6	0.774
h-5 Tetrachloroethane	1%	19.3	27.3	27.3	27.5	30.3	33.1	33.4	16.0	36.0	0.444
	2%	32.7	42.9	42.5	41.1	41.6	45.6	45.1	21.8	33.6	0.649
s : Aliphatic esters											
s-1 Ethyl acetate	1%			41.2		38.0	36.0	37.0	18.7	103.2	0.181
	2%			28.8		28.6	33.9	36.2	16.4	96.0	0.171
s-2 iso-Propyl acetate	1%	18.8	14.4	15.2	16.5	17.7	17.8	20.7	8.9	31.2	0.277
	2%	33.2	39.6	35.9	40.1	40.3	35.9	36.3	18.0	24.0	0.750
s-3 Butyl acetate	1%			30.7		32.5	31.0	33.7	16.2	91.2	0.178
	2%			35.5		36.2	37.4	38.1	18.5	79.2	0.234
s-4 iso-Amyl acetate	1%	39.5	45.1	42.6	42.6	42.6	42.4	46.4	36.3	48.0	0.756
	2%	51.2	65.9	62.4	65.0	67.1	67.1	67.2	33.6	33.6	1.000
g : Glycols and its derivatives											
g-1 Methyl cellosolve	1%	9.4	17.2	18.6	18.9	19.9	20.7	23.5	9.9	72.0	0.138
	2%	35.1	39.9	36.0	42.9	40.1	40.1	42.5	20.1	33.6	0.598
g-2 Ethyl cellosolve	1%	7.7	13.2	12.6	12.7	13.6	13.4	14.8	7.0	50.4	0.139
	2%	20.1	25.8	23.3	28.8	27.2	27.7	30.7	14.0	36.0	0.389
g-3 Butyl cellosolve	1%	17.3	18.5	20.2	24.8	22.3	35.5	25.4	11.2	40.8	0.275
	2%	24.6	35.8	37.2	39.5	41.9	43.9	43.6	21.9	48.0	0.456
g-4 Ethylene glycol	1%	41.3	56.0	49.3	49.9	48.8	45.7	46.6	24.4	24.0	1.017
	2%	36.3	46.1	44.4	46.5	46.0	50.2	49.4	23.0	40.8	0.564
g-5 Propylene glycol	1%			16.0		21.7	30.4	32.1	15.7	156.0	0.101
	2%			21.8		20.9	27.3	29.1	14.1	96.0	0.147
g-6 Butoxy propylene glycol	1%			24.9		35.2	41.2	42.6	19.0	132.0	0.144
	2%			12.6		11.6	17.6	21.7	8.5	132.0	0.064
b : Aromatic hydrocarbons and aromatic unsaturated hydrocarbons											
b-1 Benzene	1%			27.0		27.1	30.3	31.3	14.8	72.0	0.206
	2%			25.1		33.0	38.0	40.0	18.5	96.0	0.193
b-2 Monochlorobenzene	1%			16.7		17.9	22.3	22.8	10.1	96.0	0.105
	2%			38.0		34.0	36.0	38.0	18.0	72.0	0.250
b-3 Nitrobenzene	1%	34.0	42.8	40.3		43.2	43.1	39.2	21.0	31.2	0.652
	2%	38.1	46.7	45.1		46.5	50.5	43.6	22.6	19.2	1.177
b-4 Solvent naphtha	1%			28.7		32.6	32.7	36.1	16.3	120.0	0.136
	2%			35.0		34.8	37.2	36.3	18.4	72.0	0.256
b-5 Toluene	1%			30.7		28.4	30.4	33.6	14.7	96.0	0.153
	2%			24.6		31.5	34.4	35.3	17.3	96.0	0.180
b-6 o-Chlorotoluene	1%	18.3	28.5	31.4	31.0	33.4	33.5	33.4	16.7	43.2	0.387
	2%	16.4	21.6	19.1	25.6	29.7	38.0	41.0	19.8	72.0	0.275
b-7 Xylene	1%			12.0		35.8	31.2	32.1	15.8	192.0	0.083
	2%			46.7		42.2	42.6	43.0	21.9	60.0	0.365
b-8 Tri-cresol	1%	35.2	34.2	30.0	31.0	32.1	35.7	36.4	17.2	28.8	0.597
	2%	41.0	62.2	62.0	71.7	61.1	62.3	53.3	31.0	48.0	0.646

b-9 Velsicol AR-55	1%	30.6	26.5	25.5	27.5	31.0	28.8	32.2	16.0	45.6	0.350
	2%	30.3	37.0	32.1	40.3	34.7	35.2	43.0	17.5	31.2	0.561
b-10 Alpha pinene	1%	20.8	32.1	33.1	35.1	36.0	36.4	36.7	18.2	48.0	0.379
	2%	32.3	42.8	40.2	49.9	45.0	46.0	47.8	23.1	36.0	0.642
b-11 Camphor oil	1%			59.2		39.1	36.0	37.0	18.3	96.0	0.191
	2%			26.8		27.6	36.0	39.6	18.9	48.0	0.394
e: Aromatic ethers											
e-1 Dioxane	1%			29.7		30.9	31.3	32.1	15.4	84.0	0.183
	2%			32.7		32.5	34.9	36.7	12.4	72.0	0.172
Lindane 0.1%		29.25	32.61	33.16	33.16	33.16	33.16	33.16			
Carrier mixed with Clay and Talc.		1.16	1.16	1.16	1.16	1.16	3.33	7.16			

* See § Mathematical determination of the steady mortality.

Table 2. The relationship between time and per cent paralysis in the fly when carrier powders mixed with 1% and 2% solvents respectively against the adult of the housefly, *Musca domestica vicina* (the averaged values of 3 replicated trials). 28°C.

Code no. of the solvents tested	Solvent content % in w/w	Per cent paralysed fly at the elapsed time						
		1 hr.	2 hr.	3 hr.	4 hr.	5 hr.	18 hr.	24 hr.
k-1	1%			0.0		1.9	4.1	5.8
	2%			0.5		0.7	2.2	10.9
k-2	1%	0.9	1.2	1.2	1.7	0.4	0.4	0.8
	2%	1.1	1.1	1.1	1.1	1.1	3.5	4.6
k-3	1%	1.0	1.2	1.2	1.7	0.4	0.4	0.8
	2%	0.0	0.0	1.2	1.2	0.0	2.8	3.8
k-4	1%			0.0		0.0	3.8	5.2
	2%			0.0		3.5	4.6	5.8
k-5	1%	1.7	1.2	1.2	1.7	1.7	2.6	2.6
	2%	0.7	1.1	1.5	1.5	1.5	3.1	6.6
k-6	1%	0.9	1.2	1.2	1.7	0.6	0.5	1.1
	2%	0.7	1.1	1.5	1.5	1.5	1.5	3.2
k-7	1%	1.4	2.6	1.2	1.7	0.4	1.0	2.9
	2%	0.7	1.1	2.8	1.6	1.6	3.1	6.7
k-8	1%	1.7	1.2	1.2	1.7	1.7	3.3	7.2
	2%	13.4	3.7	3.7	5.3	5.3	21.7	22.8
k-9	1%	2.7	3.7	3.7	5.3	5.3	4.2	4.2
	2%	4.0	1.8	2.3	1.5	4.0	5.3	17.1
p-1	1%			0.0		0.7	1.9	2.1
	2%			0.7		0.9	3.8	8.7
p-2	1%			0.9		0.7	2.6	2.8
	2%			2.2		2.3	2.3	5.2
p-3	1%	0.8	0.8	0.8	0.8	0.8	0.8	0.8
	2%	2.2	2.2	2.2	2.2	2.2	4.4	5.5
p-4	1%			1.9		2.7	2.7	3.6
	2%			1.3		0.7	2.4	7.8
p-5	1%	1.7	1.7	1.7		1.7	1.7	6.4
	2%	1.6	2.6	3.9		6.5	7.5	8.1
p-6	1%	0.0	1.0	1.0	1.0	1.0	1.0	1.0
	2%	3.0	2.0	2.9	3.1	4.9	4.9	4.9
p-7	1%	0.7	0.7	0.7	0.7	2.0	2.7	2.7
	2%	2.3	3.4	3.0	3.0	4.1	6.0	6.7
p-8	1%	0.0	0.7	0.7	1.5	1.5	1.5	1.5
	2%	0.6	0.6	0.6	0.9	0.9	1.7	2.9
p-9	1%	0.8	0.8	1.6	2.5	2.5	2.5	2.5
	2%	3.4	3.4	3.4	4.7	4.7	5.9	7.0

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h-1	1%	8.9	2.8	2.8		3.7	5.0	5.0
	2%	6.4	5.5	5.5		7.6	10.4	11.3
h-2	1%	1.7	0.0	0.9	0.9	0.9	0.9	5.6
	2%	3.0	3.0	2.0	3.7	4.7	6.5	6.5
h-3	1%	1.3	1.3	1.3	1.3	1.3	2.1	6.6
	2%	3.4	4.6	4.6	4.6	4.6	5.8	9.4
h-4	1%	0.0	1.0	1.0	2.4	2.4	6.7	6.7
	2%	6.5	6.5	6.5	6.5	6.5	9.6	10.3
h-5	1%	3.4	4.6	4.6	3.3	0.7	0.6	1.2
	2%	4.0	8.5	5.6	12.1	12.1	8.3	18.0
s-1	1%			1.3		0.0	2.6	9.2
	2%			5.2		8.9	15.2	19.5
s-2	1%	4.8	1.9	1.9	1.9	1.9	2.9	3.9
	2%	4.3	4.3	4.3	4.3	4.3	6.5	7.8
s-3	1%			0.9		0.7	0.8	8.3
	2%			0.6		3.4	1.7	5.6
s-4	1%	4.5	6.3	6.3	4.5	1.7	3.3	7.2
	2%	6.7	8.4	10.2	10.2	11.5	14.0	14.9
g-1	1%	1.1	2.3	2.3	6.6	1.4	1.4	3.1
	2%	1.7	1.2	1.2	0.7	0.7	0.7	3.5
g-2	1%	4.6	11.0	11.0	15.8	5.1	2.7	7.4
	2%	6.6	4.6	7.0	6.6	6.6	2.3	14.8
g-3	1%	0.0	0.0	0.0	0.0	0.0	1.1	3.2
	2%	1.2	1.2	0.0	0.0	0.0	3.3	5.3
g-4	1%	1.0	1.0	2.0	2.0	2.0	2.0	2.9
	2%	3.3	5.5	5.5	6.4	6.4	6.4	8.4
g-5	1%			0.8		0.9	1.2	4.0
	2%			2.0		1.5	10.5	16.4
g-6	1%			1.0		1.0	3.7	3.7
	2%			1.7		1.3	4.1	13.1
b-1	1%			1.2		1.2	3.8	5.2
	2%			1.0		3.0	6.8	7.8
b-2	1%			1.2		3.7	3.0	6.8
	2%			1.8		4.0	6.2	12.2
b-3	1%	6.8	2.3	2.1		3.0	1.9	2.5
	2%	6.7	2.3	2.1		6.2	3.9	10.2
b-4	1%			1.0		1.0	3.0	4.0
	2%			2.4		3.5	4.7	11.4
b-5	1%			1.2		1.8	3.7	8.0
	2%			4.7		6.5	9.5	11.0
b-6	1%	2.0	0.9	0.9	0.9	0.9	3.1	3.1
	2%	0.0	1.1	1.1	1.1	1.1	3.5	5.7
b-7	1%			2.3		5.0	5.6	9.8
	2%			2.6		5.7	10.0	14.8
b-8	1%	1.5	1.2	2.3	2.3	2.3	2.3	2.3
	2%	7.2	5.0	6.3	2.4	2.4	5.9	17.8
b-9	1%	18.5	3.7	2.8	4.7	4.7	10.6	11.6
	2%	9.4	5.0	6.0	7.1	7.1	11.8	14.0
b-10	1%			0.5		0.5	1.5	2.5
	2%			25.4		22.7	22.7	28.3
e-1	1%			1.2		1.7	2.2	2.4
	2%			1.2		1.7	0.8	5.4

solvent was generally more effective than the addition of 1%. Contrary results were sometimes obtained because it seems that the solvents of

higher viscosity such as glycols interfere with the cuticular penetration of lindane.

In the case of the addition of 1% solvent, the

higher carrier efficiencies of the solvents, in descending order of effectiveness, were as follows: 1) ethylene glycol 2) chloroform 3) *iso*-butylalcohol 4) *iso*-amyl acetate 5) nitrobenzene 6) ethylene dichloride 7) cyclohexanol 8) trichloroethylene 9) *n*-propyl alcohol 10) butoxy propylene glycol.

In the case of the addition of 2% solvent, the higher efficiencies were expressed as follows: 1) *iso*-butyl alcohol 2) cyclohexanol 3) methyl *iso*-butyl ketone 4) chloroform 5) *iso*-amyl acetate 6) tri-cresol 7) ethylene dichloride and *n*-butyl alcohol 9) trichloroethylene 10) benzyl alcohol, ethylene glycol, acetophenone and *n*-propyl alcohol.

On adding the values of order obtained with both 1% and 2% of solvent, the following descending order of effectiveness was obtained: 1) chloroform 2) *iso*-butyl alcohol 3) ethylene glycol 4) methylethyl ketone 5) ethylene dichloride 6) cyclohexanol 7) *iso*-amyl acetate 8) nitrobenzene 9) *n*-propyl alcohol 10) cyclohexanone.

The above expression of order was obtained from the asymptote of the time and per cent paralysed-fly-curve which was termed the steady mortality.

Mathematical determination of steady mortality

As shown in Figure 1 the accumulated curve representing the relation between time and per cent paralysis is a sigmoid curve (a-b-d curve such as Figure 1) or an a-c-b-d curve such as Figure 1.

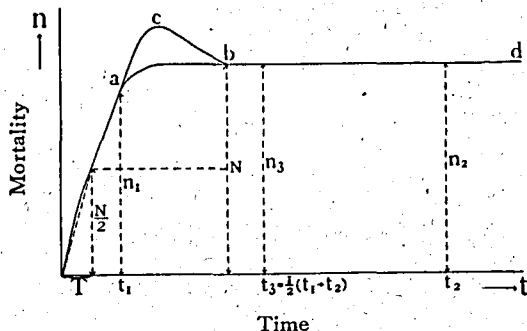


Fig. 1. Schematic graph showing the accumulated curve indicated the relation between time and per cent paralysis in the fly.

For convenience, we regard the a-c-b-d curve of narcotic solvent as an a-b-d curve in the present paper. The b-d part of the curve is the part of steady mortality.

Although the data analysis of the present curves were calculated by the calculation of the incomplete time-mortality curve of Bliss⁹⁾ a short cut method was applied as follows:

We applied the modification of Kono's method¹⁸⁾ on the estimation of insect population by time-unit-collecting.

In the experiment on the relation between time and per cent mortality, the relation between the elapsed time t , and the accumulated mortality at time t , n can be represented by the following equation:

$$n = N(1 - e^{-at}) \quad (1)$$

where a is a constant representing the mortality efficiency, N the steady mortality in a given toxicant, which can be calculated by the following equation, derived from the equation (1):

$$N = \frac{n_1 n_2 - n_3^2}{n_1 + n_2 - 2n_3} \quad (2)$$

where n_1 , n_2 and n_3 are the mortality at the time, t_1 , t_2 and $t_3 = 1/2(t_1 + t_2)$, respectively.

$n = N/2$, dn/dt becomes the maximum velocity, thus when the steady mortality is regarded as 100%, $N/2$ becomes the median lethal time and then is the point of inflection.

In the present case, N , was calculated by equation (1) and $N/2$ was obtained. The index representing the slope of a given curve was calculated as follows:

$$\text{The index of the slope of curve} = \frac{N}{2T} \quad (3)$$

where T is time, t , at $N/2$.

The index of the slope is an important factor in the cuticular penetration but the values of the index for the present experiments were not more distinct than the values of the steady mortality.

As we thought that the solvents with a higher steady mortality value have the greater probability to reach the action point of insect body by the greater carrier efficiency, we took in the values of the steady mortality as important factor on the cuticular penetration.

The effects of solvents on cuticular permeability of lindane against the oriental garden cricket

The results of the experiments for the effects of solvents on cuticular permeability of lindane against the oriental garden cricket are given in Table 3.

The results with the cricket differed from the

Table 3. The time in minutes at which the oriental garden crickets paralyse after treatment with the mixture of 0.1% lindane and 99.9% solvents. The tarsus of the cricket's hind leg was immersed into the test solution for 30 seconds (the averaged values of 3 replicated trials). 38°C.

Code no. of Solvent tested	Time in minutes at which crickets paralyse after immersion with the mixture.	
	Female	Male
k-1	345	62
k-2	51	51
k-3	366	250
k-4	32	41
k-5	100	44
k-6	29	49
k-7	30	29
k-8	46	23
k-9	26	4
p-1	31	33
p-2	83	255
p-3	48	123
p-4	75	125
p-5	298	179
p-6	149	26
p-7	115	31
p-8	1,978	36
p-9	21	19
p-10*	24	36
p-11*	61	62
h-1	60	57
h-2	192	416
h-3	182	30
h-4	18	48
h-5	31	358
s-1	256	198
s-2	91	68
s-3	63	248
s-4	18	5
g-1	541	85
g-2	1,068	2,779
g-3	29	36
g-4	148	238

results with the housefly in descending order of carrier efficiency to the solvents.

In the case of the cricket, we observed that the time taken for cricket paralysis varied with the kind of organic solvent. Usually paralysis was more rapid with the female than with the male.

In the case of the male cricket, the higher efficiencies of the solvents, in descending order of effectiveness, were as follows: 1) benzene 2) methyl *iso*-butyl ketone and aniline 4) *iso*-amyl acetate 5) dioxane 6) *n*-hexane 7) cyclohexanol and toluene 9) *o*-chlorotoluene 10) methyl ketone.

In the case of the female cricket, the higher efficiencies were as follows: 1) Velsicol AR-60 (methyl naphthalene oils) 2) toluene 3) *iso*-amyl acetate, dioxane and trichloroethylene 6) *o*-chloro-

g-5	197	169
g-6	187	540
b-1	118	1
b-2	33	96
b-3	30	87
b-4	20	85
b-5	16	19
b-6	19	22
b-7	39	43
b-8	515	42
b-9	65	42
b-10	57	31
b-11	24	235
b-12*	52	54
b-13*	3	33
b-14*	58	64
b-15*	21	39
b-16*	27	11
e-1	18	7
a : Amines		
a-1	48	4
a-2	45	26
Kerosene, k-1	25	48
Dimethyl phthalate, d-1	298	179
o : Plant oils		
o-1*	95	146
o-2*	417	415

* p-10 : octyl alcohol, p-11 : decyl alcohol, b-12 : Velsicol AR-50, b-13 : Velsicol AR-60, b-14 : methyl naphthalene oil, b-15 : Penn Drake oil, b-16 : *n*-hexane, a-1 : aniline, a-2 : dimethyl aniline, o-1 : peanut oil, o-2 : sesame oil.

rotoluene 7) solvent naphtha 8) cyclohexanol and Penn Drake oils (methyl naphthalene oils) 10) *n*-octyl alcohol.

On adding the values of order obtained with both the male and female, 1) *iso*-amyl acetate 2) dioxane 3) toluene 4) *o*-chlorotoluene 5) cyclohexanol 6) methyl *iso*-butyl ketone 7) Velsicol AR-60 8) *n*-hexane 9) Penn Drake oils 10) trichloroethylene.

Combining the values obtained for the cricket with that of the housefly resulted in the carrier efficiencies of the solvents, the following descending order of effectiveness was obtained: 1) *iso*-amyl acetate 2) cyclohexanol 3) trichloroethylene 4) methylethyl ketone 5) cyclohexanone and methyl *iso*-butyl ketone 7) *iso*-butyl alcohol, *o*-chlorotoluene and *n*-hexane 10) dioxane 11) di-*iso*-butyl ketone and butyl cellosolve 13) nitrobenzene 14) solvent naphtha and diethyl ketone 16) chloroform and toluene 18) *iso*-amyl alcohol and ethylene dichloride 20) acetophenone 21) xylene 22) camphor oil 23) *n*-propyl alcohol 24) ethyl alcohol 25) methyl alcohol 26) ethylene glycol 27) tricresol 28) alpha-pinene and benzene 30) tetrachloroethane 31) benzyl alcohol 32) butyl acetate and acetone 34) *n*-butyl alcohol and monochlorobenzene 36) *iso*-propyl acetate 37) methyl naphthalene oil 38) carbon tetrachloride 39) diacetone alcohol and *iso*-propyl alcohol 41) ethyl acetate 42) butoxy propylene glycol 43) acetonyl acetone, 44) ethyl cellosolve.

Considering the results obtained in the overall experiments with the flies and crickets on aliphatic ketones, aliphatic alcohols, halogenated hydrocarbons, glycols and its derivatives, the esters of acetic acid and the derivatives of benzene, it was found that the derivatives of benzene such as *o*-chlorotoluene and benzene had the highest efficiency. Aliphatic ketones such as methylethyl ketone and halogenated hydrocarbons such as trichloroethylene had moderate efficiencies.

Generally speaking, glycol and its derivatives, the esters of acetic acid and aliphatic alcohols had low efficiencies. In the individual case of the cricket, the carrier efficiencies of the above groups tested, in descending order of effectiveness, were expressed as follows: 1) the derivatives of benzene 2) halogenated hydrocarbons. 3) aliphatic ketones 4) the esters of acetic acid 5) aliphatic alco-

hol 6) glycols and its derivatives. In the individual case of the housefly, the order was as follows: 1) halogenated hydrocarbons 2) the esters of acetic acid and aliphatic alcohols 4) aliphatic ketones 5) the derivatives of benzene 6) glycols and its derivatives.

Relationship between carrier efficiency and boiling point of solvents

It has been stated that there is a general relationship between the toxicity of a poison and its boiling point by numerous investigators. We attempted to find the tendency in a general relationship between the carrier efficiency and the boiling point of the solvent.

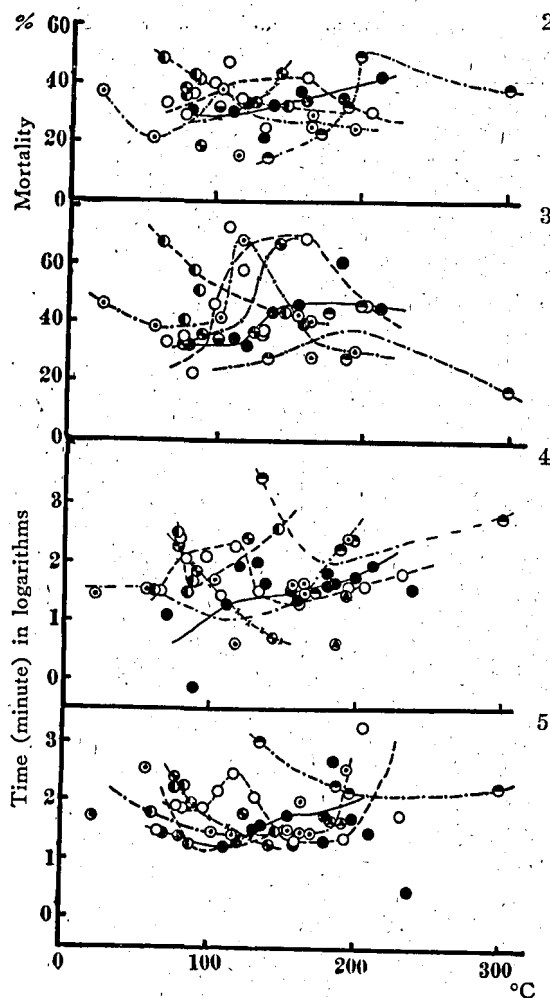
According to Webb & Green³⁰, the solvents with comparatively high volatility have the lower carrier efficiency because such solvents reduce the additional volume in the active dust by rapid evaporation of solvent and solvents with a comparatively high volatility would evaporate from the cuticle surface, then would lose carrier efficiency once their volume fell below that necessary to maintain the insecticide in solution.

The considerations of the experimental results on this relationship are indicated in Table 4 and in Figures 2 to 5.

As shown in the case of the derivatives of benzene, the solvents with the lower volatility beyond 156° had moderate steady mortalities in the experiment on the effect of 2% dusts on the housefly and the results with 1% dusts indicated that the steady mortality increased from about 120°. In the case of the male cricket, the speed of paralysis gradually reduced with the increase of boiling point. In the case of the female cricket, the relationship between the logarithmic value of the speed of paralysis and the boiling point are indicated by a line curved like a bowl, thus the effective range stood about 110°. In the case of action of 1% and 2% dusts to the housefly, aliphatic ketones were the curves with the peak of 102.7° and 117°, respectively. Aliphatic primary alcohols indicated the typical curves with the maximum effective range from 120 to 160°. In addition of 1% and 2% dusts to the halogenated hydrocarbons, the carrier efficiency gradually reduced with the

Table 4. The relationship between carrier efficiency and boiling point of solvents against the oriental garden cricket and the housefly (3 replicated trials).

Solvent tested	The boiling point of solvent °C	Logarithmic values transformed the time of paralysis to the cricket		The steady mortality of the housefly	
		Female	Male	1%	2%
Acetophenone	20.5	1.7076	1.7076	35.5	46.0
Acetone	56.24	2.5378	1.7924	20.0	38.7
Chloroform	61.2	1.7782	1.7559	47.2	67.2
Methyl alcohol	64.7	1.4914	1.5185	32.0	33.0
<i>n</i> -Hexane	69.0	1.4314	1.0414		
Carbon tetrachloride	76.8	2.2833	2.5191	36.0	40.3
Ethyl acetate	77.1	2.4082	2.2967	37.4	32.7
Ethyl alcohol	78.4	1.9191	2.4065	28.5	35.0
Kerosene	80.0	1.3979	1.6812		
Benzene	80.1	2.0719	0.0000	29.6	37.0
<i>iso</i> -Propyl alcohol	82.5	1.8751	2.0969	35.2	32.3
Ethylene dichloride	83.5	2.2601	1.4771	41.9	58.0
Trichloroethylene	87.2	1.2553	1.6812	40.3	51.9
<i>iso</i> -Propyl acetate	89.0	1.9590	1.8325	17.7	35.9
<i>n</i> -Propyl alcohol	97.0	1.8751	2.0969	38.8	46.0
Dioxane	100.8	1.2553	0.8451	30.8	34.7
Diethyl ketone	102.7	1.4624	1.6902	36.7	40.3
<i>iso</i> -Butyl alcohol	109.0	2.1732	1.4150	46.0	73.0
Toluene	110.6	1.2041	1.2788	29.4	34.5
<i>n</i> -Butyl alcohol	117.0	2.4742	2.2529	34.0	58.0
Methyl- <i>iso</i> -butyl ketone	117.0	1.4150	0.6021	14.2	68.0
Solvent naphtha	120.0	1.3010	1.9294	32.6	36.7
Butyl acetate	125.1	1.7993	2.3945	32.4	37.0
Monochlorobenzene	131.7	1.5185	1.9823	20.2	36.0
<i>iso</i> -Amyl alcohol	132.0	2.0607	1.4914	23.3	36.0
Ethyl cellosolve	134.5	3.0286	3.4439	13.9	27.9
Xylene	137.0	1.5911	1.6335	31.6	43.7
<i>iso</i> -Amyl acetate	142.0	1.2553	0.6990	42.6	67.1
Tetrachloroethane	146.3	1.4914	2.5539	32.0	43.6
Alpha pinene	155.0	1.7559	1.4914	36.4	46.2
Cyclohexanone	155.0	1.5051	1.6128	36.2	42.8
<i>o</i> -Chlorotoluene	159.5	1.2788	1.3424	33.4	39.5
Cyclohexanol	160.0	1.3222	1.2788	41.1	69.0
Diacetone alcohol	164.0	2.0000	1.6435	24.8	28.0
Di- <i>iso</i> -butyl ketone	164.0	1.4771	1.4624	28.1	40.8
Butyl cellosolve	170.6	1.4624	1.4771	22.3	43.7
Methyl naphthalene oil	180.0	1.7634	1.8062		
Penn Drake oil	180.0	1.3222	1.5911		
Aniline	184.4	1.6812	0.6021		
Tri-cresol	185.0	2.7118	1.6232	34.3	61.9
Propylene glycol	188.0	2.2945	2.2279	31.3	28.2
Dimethyl aniline	192.0	1.6532	1.4150		
Acetonylacetone	194.0	2.5635	2.3979	24.1	30.5
Octyl alcohol	194.0	1.3802	1.5562		
Ethylene glycol	197.2	2.1703	2.3766	48.8	46.0
Velsicol AR-50	199.0	1.7160	1.7324		
Benzyl alcohol	205.2	3.2956	1.5563	29.6	46.0
Nitrobenzene	210.9	1.4771	1.9395	42.0	45.1
Decyl alcohol	232.0	1.7853	1.7924		
Velsicol AR-60	238.0	0.4771	1.5185		
Butoxy propylene glycol	300.0	2.2719	2.7324	38.0	17.0



Figs. 2 to 5. Relation between carrier efficiency and the boiling point of solvents against the housefly (Figs. 2-3) and the oriental garden cricket (Figs. 4-5).

Y-axis represents the steady mortality in percentage at Figs. 2-3, and the logarithmic values transformed the time in minute of paralysis at Figs. 4-5. X-axis represents the boiling point of solvents.

Fig. 2 : The addition of 1% solvent against the housefly.

Fig. 3 : The addition of 2% solvent against the housefly.

Fig. 4 : The male of the cricket. Fig. 5 : The female of the cricket.

—○— Primary alcohols —●— Halogenated hydrocarbons —●— Aromatic hydrocarbons —●— Aromatic ethers —●— Glycols and its derivatives —●— Esters —●— Aliphatic, aromatic and cyclic ketones —▲— Anilines

2 progress of boiling point.

The relationship between carrier efficiency and boiling point in the other solvents was given in Table 4 and in Figures 2 to 5.

At a glance, the lines to both sexes were generally indicated the line curved like a bowl with the exception of some solvent-groups as shown in Figures 4 to 5.

Relationship between carrier efficiency and solubility of solvents to lindane.

The relationship between carrier efficiency and the solubility of solvents to lindane is given in table 5

We could not recognized a tendency to a general relationship between carrier efficiency and the solubility of solvents to lindane. Whereas the solubility of solvents to lindane involves the governable factors concerned in a practical formulation, the lindane-solubility did not indicate a clear relationship in the present experiments. Although there were many unfavourable results in this phase of the work, it seems that with the primary alcohols there was a slight relationship in the case of the housefly. In the experiments with the housefly, the carrier efficiencies of the derivatives of ketone gradually increased with the increase in the solubility of lindane. Such relationships however, were not clear in the case of the cricket.

Relationship between carrier efficiency and the solubility of solvents to beeswax.

It is well known that the cuticle of an insect is essentially a two phase system of wax and water with a definite interface between the two layers. It may be supposed that the outer protective wax layer removed by solvents causes increased susceptibility to insecticides.

Starting with this conception, we attempted to find a relationship between carrier efficiency and solubility of solvents to beeswax. The results with the beeswax showed a similar tendency as

Table 5. The relationship between the solubility of solvents to lindane and the carrier efficiency of solvents against the oriental garden cricket and the housefly (3 repeated tests).

Solvent tested	The solubility to lindane % in w/w 30°C	The logarithmic values transformed the time of paralysis against the cricket		The steady mortality of the housefly	
		Female	Male	1%	2%
Acetone	59.1	2.5378	1.7924	20.0	38.7
Methyl ethyl ketone	56.6	1.6628	1.3617	43.8	64.9
Diethyl ketone	53.2	1.4624	1.6902	36.7	40.3
Cyclohexanone	52.7	1.5050	1.6128	36.2	42.8
Ethyl acetate	50.4	2.4082	2.2967	37.4	32.7
Acetophenone	45.3	1.7076	1.7076	35.5	46.0
Methyl- <i>iso</i> -butyl ketone	42.4	1.4150	0.6021	14.2	68.0
Benzene	42.3	2.0719	0.0000	29.6	37.0
Butyl acetate	41.6	1.7993	2.3945	32.4	37.0
Acetonylacetone	39.5	2.5635	2.3979	24.1	30.0
<i>iso</i> -Propyl acetate	38.9	1.9590	1.8325	17.7	35.9
Toluene	38.0	1.2041	1.2788	29.4	34.5
Nitrobenzene	36.7	1.4771	1.9395	42.0	45.1
<i>iso</i> -Amyl acetate	34.0	1.2553	0.6990	42.5	67.1
Xylene	32.7	1.5911	1.6335	31.6	43.7
Monochlorobenzene	31.9	1.5185	1.9823	20.2	36.0
<i>o</i> -Chlorotoluene	30.2	1.2788	1.3424	33.4	39.5
Di- <i>iso</i> -butyl ketone	27.0	1.4771	1.4624	28.1	40.8
Chloroform	23.5	1.7782	1.7559	47.2	67.2
Tetrachloroethane	22.0	1.4914	2.5539	32.0	43.0
Methyl cellosolve	21.2	2.7332	1.9294	19.8	40.1
Ethyl cellosolve	21.0	3.0286	3.4439	13.9	27.9
Di acetone alcohol	20.1	2.0000	1.6435	24.8	28.0
Butyl cellosolve	19.4	2.1732	1.4150	22.3	43.7
<i>n</i> -Butyl alcohol	5.6	2.4742	2.2529	34.0	56.0
Cyclohexanol	5.4	1.3222	1.2788	41.1	69.0
Methyl alcohol	5.2	1.4914	1.5185	32.0	33.0
<i>n</i> -Propyl alcohol	5.0	1.6812	2.0899	38.8	46.0
<i>iso</i> -Propyl alcohol	3.7	1.8751	2.0669	35.2	32.3
Penn Drake oil	3.2	1.3222	1.5911		
Ethyl alcohol	2.8	1.9191	2.4065	28.5	35.0

the relation to the solubility of lindane.

The relationship between carrier efficiency and the solubility of solvents to beeswax is given in Table 6.

Beeswax has the general property of the lipoids of the insect cuticle but the chemical and physical properties of beeswax in detail differ with the waxes of the housefly and the cricket. The solubility of beeswax did not show a clear relationship. The unfavourable results of the relation to beeswax were probably resulted from the difference between the beeswax and the insect's waxes tested.

Webb & Green³⁰⁾ pointed out that the ability to penetrate wax, though essential to a solvent

showing high carrier efficiency, is not the only factor involved.

In the case of the present investigation, five solvents showing comparatively high carrier efficiencies such as trichloroethylene, *n*-hexano, cyclohexanol, cyclohexanone and *o*-chlorotoluene showed higher solvent action on beeswax.

Relationship between carrier efficiency and the solubility of solvents to the housefly's lipoids

The results on the action of solvents on the housefly's lipoids were given in Table 7. In this case, the solubility of housefly's lipoids did not indicate a clear relationship. However, the solvents with the higher solvency such as tri-

Table 6. The relationship between carrier efficiency and the solubility of solvents to beeswax against the oriental garden cricket and the housefly (3 replicated trials).

Solvent tested	The solubility of solvents to beeswax % in w/w, 30°C	Log. values transformed the time of paralysis to the cricket		The steady mortality of the housefly	
		Female	Male	1%	2%
Methyl cellosolve	0.075	2.7332	1.9294	19.8	40.1
Methyl alcohol	0.111	1.4114	1.5185	32.0	33.0
Ethylene glycol	0.161	2.1703	2.3766	48.8	46.0
Aniline	0.162	1.6812	0.6021		
Acetonylacetone	0.195	2.5635	2.3979	24.1	30.5
Methyl ethyl ketone	0.199	1.6628	1.3617	43.8	64.9
Ethyl cellosolve	0.217	3.0286	3.4439	13.9	27.9
Dimethyl phthalate	0.231	2.4742	2.2529		
Ethyl alcohol	0.313	1.9191	2.4065	28.5	35.0
Penn Drake oil	0.335	1.3222	1.5911		
Dioxane	0.594	1.2553	0.8451	30.8	34.7
Acetophenone	0.850	1.7076	1.7076	35.5	46.0
<i>n</i> -Propyl alcohol	0.931	1.6812	2.0899	38.8	46.0
Butyl cellosolve	0.963	1.4624	1.4771	22.3	43.7
<i>iso</i> -Butyl alcohol	0.994	2.1732	1.4150	46.0	73.0
<i>n</i> -Butyl alcohol	1.006	2.4742	2.2529	34.0	56.0
<i>iso</i> -Amyl alcohol	1.247	2.0607	1.4914	23.3	36.0
Propylene glycol	1.295	2.2945	2.2279	31.3	28.2
Methyl- <i>iso</i> -butyl ketone	1.323	1.4150	0.6021	14.2	68.0
<i>iso</i> -Propyl alcohol	1.367	1.8751	2.0969	35.2	32.3
Butoxy propylene glycol	1.499	2.2718	2.7324	38.0	17.0
Butyl acetate	1.571	1.7993	2.3945	32.4	37.0
<i>iso</i> -Amyl acetate	2.052	1.2553	0.6990	42.6	67.1
<i>iso</i> -Propyl acetate	2.775	1.9590	1.8325	17.7	35.9
Dimethylaniline	2.979	1.6532	1.4150		
Kerosene	3.056	1.3979	1.6812		
Diethyl ketone	3.229	1.4624	1.6902	36.7	40.3
Nitrobenzene	3.438	1.4771	1.9395	42.0	45.1
Ethylene dichloride	3.472	2.2601	1.4771	41.9	58.0
Di- <i>iso</i> -butyl ketone	3.764	1.4771	1.4624	28.1	40.8
Solvent naphtha	4.571	1.3010	1.9294	32.6	36.7
Velsicol AR-50	5.460	1.7160	1.7324		
Tetrachloroethylene	5.896	1.4914	2.5539	32.0	43.0
Methyl naphthalene oil	5.908	1.7634	1.8062		
Alpha pinene	6.123	1.7559	1.4914	36.4	46.2
Acetone	6.240	2.5378	1.7924	20.0	38.7
<i>n</i> -Hexane	6.742	1.4314	1.0414		
<i>o</i> -Chlorotoluene	7.064	1.2788	1.3424	33.4	39.4
Cyclohexanone	7.180	1.5051	1.6128	36.2	42.8
Cyclohexanol	7.674	1.3222	1.2788	41.1	69.0
Carbon tetrachloride	8.469	2.2833	2.6191	31.6	43.7
Xylene	8.731	1.5911	1.6335	20.2	36.0
Monochlorobenzene	9.703	1.5185	1.9823	29.6	37.0
Benzene	13.528	2.0719	0.0000	29.4	34.5
Toluene	15.361	1.2041	1.2788	40.3	51.9
Trichloroethylene	17.052	1.2553	1.6812	47.2	67.2
Chloroform	18.171	1.7782	1.7559	29.6	46.0
Benzyl alcohol	19.660	3.2956	1.5563	24.8	28.0
Di-acetone alcohol	24.296	2.0000	1.6435	36.0	40.3

Table 7. The speed of solubility of solvents to beeswax and the housefly's lipoids. Approx. 0.02g of Beeswax and 0.006g of the fly lipoids were examined to dissolve into 5cc of solvents at 30°.

Code no. of solvent tested	Time in seconds at which one drop of the fly lipoids dissolve into solvents	Time in hours at which one drop of beeswax dissolve into solvents
k-1	above 24 hr.	above 48.000
k-2	78.8	above 48.000
k-3	above 24 hr.	above 48.000
k-4	86.0	24.000
k-5	above 24 hr.	above 48.000
k-6	92.8	38.000
k-7	117.3	4.667
k-8	89.6	above 48.000
k-9	205.6	36.000
p-1	above 24 hr.	above 48.000
p-2	above 24 hr.	above 48.000
p-3	above 24 hr.	above 48.000
p-4	above 24 hr.	above 48.000
p-5	20.2	above 48.000
p-6	above 24 hr.	above 48.000
p-7	above 24 hr.	above 48.000
p-8	below 24 hr.	above 48.000
p-9	136.0	above 48.000
p-10	above 24 hr.	
p-11	below 24 hr.	
h-1	15.8	0.050
h-2	55.0	0.083
h-3	87.0	1.500
h-4	71.0	0.083
h-5	67.0	0.167
s-1	above 24 hr.	36.000
s-2	100.8	above 48.000
s-3	111.6	36.000
s-4	292.0	36.000
g-1	above 24 hr.	above 48.000
g-2	above 24 hr.	above 48.000
g-3	101.6	above 48.000
g-4	430.0	above 48.000
g-5	above 24 hr.	above 48.000
g-6	below 24 hr.	above 48.000
b-1	below 24 hr.	0.167
b-2	66.0	0.167
b-3	137.0	above 48.000
b-4	172.5	0.167
b-5	below 24 hr.	0.083
b-6	88.0	0.083
b-7	98.4	0.083
b-8	above 24 hr.	36.000
b-9	below 24 hr.	0.667
b-10	172.2	0.916
b-11	130.5	0.500

b-12	below 24 hr.	0.916
b-13	below 24 hr.	
b-14	108.5	0.916
b-15	below 24 hr.	12.000
b-16	below 24 hr.	0.333
c-1	below 24 hr.	above 48.000
a-1	100.4	above 48.000
a-2	115.0	0.833
k-1	94.6	1.283
d-1	above 24 hr.	above 48.000
o-1	above 24 hr.	above 48.000
o-2	above 24 hr.	above 48.000

chloroethylene, cyclohexanone, *o*-chlorotoluene, di-*iso*-butyl ketone, butyl cellosolve and chloroform had comparatively higher carrier efficiency than the other solvents.

Relationship between carrier efficiency and surface tension of solvents

The surface tension of solvents is an important factor in the cuticular penetration of insecticides. Any reduction of the surface tension will increase the ability of the solvent to spread and wet the insect cuticle.

The relationship between the surface tension of solvents saturated with lindane and carrier efficiency is given in Table 8 and in Figures 6 to 9.

As shown in Table 8, the relationship between carrier efficiency and the surface tension against the crickets was more obvious than the relationship against the housefly. There were however, many unfavourable results with both insects.

In the case of the female cricket, the speed of paralysis gradually reduced with an increase of the surface tension of the aliphatic primary alcohols, aliphatic ketones and aromatic hydrocarbons (the derivatives of benzene). In the case of the male cricket, similar results to that female were obtained with the aliphatic ketones, aromatic hydrocarbons, the esters of acetic acid and aliphatic alcohols.

Whereas there were the slight relationships between carrier efficiency and surface tension with aliphatic ketones and halogenated hydrocarbons to 2% dusts and with only the aliphatic

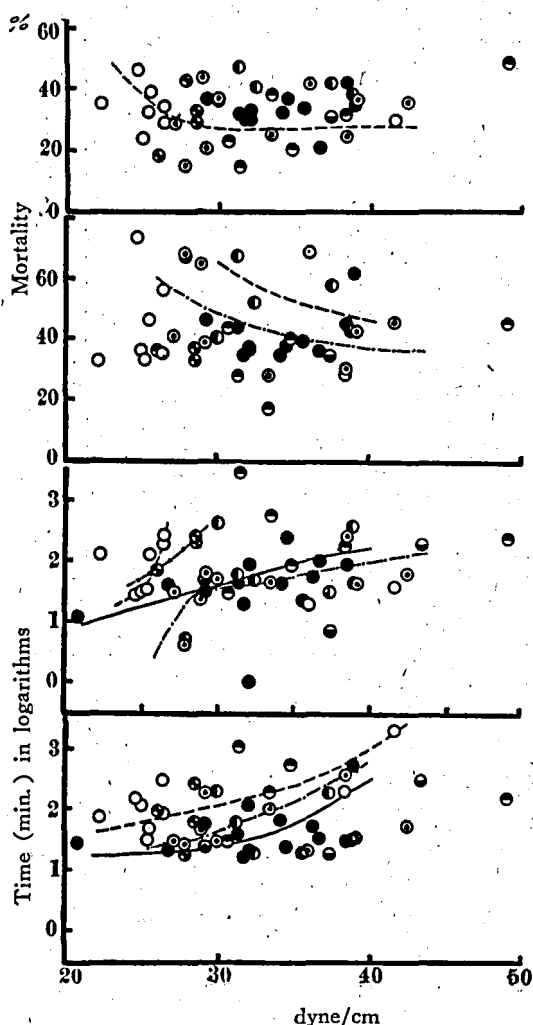
Table 8. The relationship between carrier efficiency and the surface tension of solvents saturated with lindane against the oriental garden cricket and the housefly (3 replicated trials).

Solvent tested	The surface tension of solvents, 30°C (dyne/cm)	Log. values transformed the time of paralysis to the cricket		The steady mortality of the housefly	
		Female	Male	1%	2%
<i>n</i> -Hexane	20.64	1.4314	1.0414		
<i>iso</i> -Propyl alcohol	22.14	1.8751	2.0969	35.2	32.3
<i>iso</i> -Butyl alcohol	24.56	2.1732	1.4150	46.0	73.0
<i>iso</i> -Amyl alcohol	24.91	2.0607	1.4914	23.3	36.0
Methyl alcohol	25.27	1.4914	1.5185	32.0	33.0
<i>n</i> -Propyl alcohol	25.41	1.6812	2.0899	38.8	46.0
<i>iso</i> -Propyl acetate	25.98	1.9590	1.8325	17.7	35.9
Ethyl alcohol	26.34	1.9191	2.4065	28.5	35.0
<i>n</i> -Butyl alcohol	26.34	2.4742	2.2529	34.0	56.0
Penn Drake oil	26.69	1.3222	1.5911		
Di- <i>iso</i> -butyl ketone	27.05	1.4771	1.4624	28.1	40.8
Methyl <i>iso</i> -butyl ketone	27.76	1.4150	0.6021	14.2	68.0
<i>iso</i> -Amyl acetate	27.76	1.2553	0.6990	42.5	67.1
Ethyl acetate	28.47	2.4082	2.2967	37.4	32.7
Butyl acetate	28.47	1.7993	2.3945	32.4	37.0
Methylethyl ketone	28.83	1.6628	1.3617	43.8	64.9
Kerosene	29.18	1.3979	1.6812		
Alpha pinene	29.18	1.7559	1.4914	36.4	46.2
Acetone	29.18	2.5378	1.7924	20.0	38.7
Carbon tetrachloride	29.90	2.2833	2.6191	36.0	40.3
Diethyl ketone	29.90	1.4624	1.6902	36.7	40.3
Butyl cellosolve	30.61	1.4624	1.4771	22.3	43.7
Chloroform	31.25	1.7782	1.7559	47.2	67.2
Xylene	31.32	1.5911	1.6335	31.6	43.7
Ethyl cellosolve	31.39	3.0286	3.4439	13.9	27.9
Toluene	31.68	1.2041	1.2788	29.4	34.5
Solvent naphtha	32.03	1.3010	1.9294	32.6	36.7
Benzene	32.03	2.0719	0.0000	29.6	37.0
Trichloroethylene	32.39	1.2553	1.6812	40.3	51.9
Diacetone alcohol	33.45	2.0000	1.6435	24.8	28.0
Butoxy propylene glycol	33.45	2.2718	2.7324	38.6	17.0
Velsicol AR-55	34.17	1.8129	1.6232	32.0	34.9
Camphor oil	34.52	1.3802	2.3711	36.5	37.8
Methyl cellosolve	34.81	2.7332	1.9294	19.8	40.1
<i>o</i> -Chlorotoluene	35.59	1.2788	1.3423	33.4	39.5
Cyclohexanol	35.95	1.3222	1.2788	41.1	69.0
Velsicol AR-50	36.23	1.7160	1.7324		
Monochlorobenzene	36.66	1.5185	1.9823	20.2	36.0
Ethylene dichloride	37.37	2.2601	1.4771	41.9	58.0
Dioxane	37.37	1.2553	0.8451	30.8	34.7
Propylene glycol	38.37	2.2945	2.2279	31.3	28.2
Nitrobenzene	38.44	1.4771	1.9395	42.0	45.1
Acetylacetone	38.44	2.5635	2.3979	24.1	30.5
Tetrachloroethane	38.79	1.4914	2.5539	38.0	43.0
Tri-cresol	38.94	2.7118	1.6232	34.3	61.8
Cyclohexanone	39.15	1.5051	1.6128	36.2	42.8
Benzyl alcohol	41.64	3.2956	1.5563	29.6	46.0
Acetophenone	42.49	1.7076	1.7076	35.5	46.0
Dimethyl phthalate	43.42	2.4742	2.2529		
Ethylene glycol	49.11	2.1703	2.3766	48.8	46.0

alcohols to 1% dusts against the housefly, the surface tensions to the other solvents did not generally indicate a clear relationships.

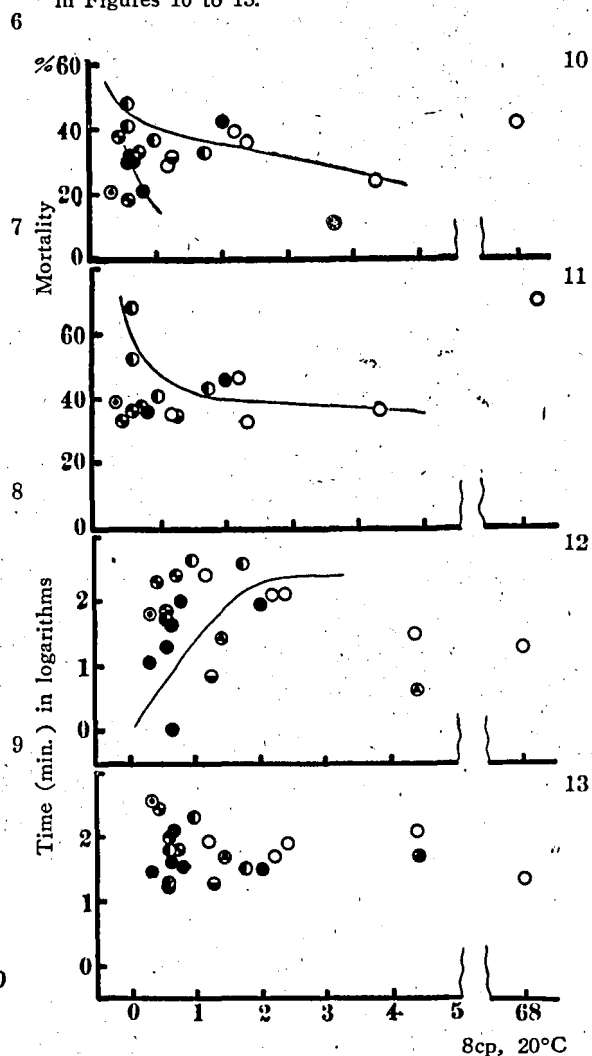
Relationship between carrier efficiency and viscosity of solvents

The rate of spreading is dependent upon the



Figs. 6 to 9. Relation between carrier efficiency and the surface tension of solvents saturated with lindane against the housefly and the oriental garden cricket. X-axis represents the surface tension of solvents saturated with lindane. Figs. 6 and 7 indicate the addition of 1% and 2% solvent, respectively against the fly. Figs. 8 and 9 indicate the male and the female of the cricket respectively.

viscosity of the solvents. The viscosity of the solvents bears an inverse relationship to the surface tension. Accordingly, the viscosity of solvents is an important factor concerned in cuticular penetration. The relationship between the viscosity of solvents and carrier efficiency at a temperature of 20° are shown in Table 9 and in Figures 10 to 13.



Figs. 10 to 13. Relation between carrier efficiency and the viscosity of solvents against the housefly and oriental garden cricket. X-axis represents the viscosity of solvents (cp) under the temperature of 20°. Figs. 10 and 11 represent the addition of 1% and 2% solvent against the fly respectively. Figs. 12 and 13 represent the male and the female of the cricket respectively.

Table 9. The relationship between carrier efficiency and the viscosity of solvents against the housefly and the oriental garden cricket (3 replicated trials).

Solvent tested	The viscosity of solvent, 20°C (cp)	Log. values transformed the time of paralysis to the cricket		The steady mortality of the housefly	
		Female	Male	1%	2%
<i>n</i> -Hexane	0.320	1.4314	1.0414		
Acetone	0.322	2.5378	1.7924	20.0	38.7
Ethyl acetate	0.449	2.4082	2.2967	37.4	32.7
Chloroform	0.570	1.7782	1.7559	47.2	67.2
Propyl acetate	0.580	1.9590	1.8325	17.7	35.9
Trichloroethylene	0.580	1.2553	1.6812	40.3	51.9
Toluene	0.586	1.2041	1.2788	29.4	34.5
Xylene	0.610	1.5911	1.6335	31.6	43.7
Benzene	0.650	2.0719	0.0000	29.6	37.0
Butyl acetate	0.732	1.7993	2.3945	32.4	37.0
Monochlorobenzene	0.800	1.5185	1.9823	20.2	36.0
Carbon tetrachloride	0.970	2.2833	2.6191	36.0	40.3
Ethyl alcohol	1.190	1.9191	2.4065	28.5	35.0
Dioxane	1.260	1.2553	1.8451	30.8	34.7
Dimethyl aniline	1.410	1.6532	1.4150		
Tetrachloroethane	1.750	1.4914	2.5539	32.0	43.0
Nitrobenzene	2.010	1.4771	1.9395	42.0	45.1
<i>n</i> -Propyl alcohol	2.200	1.6812	2.0899	38.8	46.0
<i>iso</i> -Propyl alcohol	2.390	1.8751	2.0969	35.2	32.3
<i>iso</i> -Amyl alcohol	4.360	2.0607	1.4914	23.3	36.0
Aniline	4.400	1.6812	0.6021		
Cyclohexanol	68.000	1.3222	1.2788	41.1	69.0

The results with the male cricket and the housefly treated with 1% dusts show a fairly clear relationship to the viscosity.

The carrier efficiency gradually reduced with an increase in viscosity. The results with the housefly treated with a 2% dust indicated a similar tendency as the 1% dust with the exception of some groups of solvents. In the case of the female cricket, the viscosity of solvents did not indicated any clear relationship.

Relationship between carrier efficiency and some limiting factors governing the solubility of solvents

As shown by Kuwada¹⁰⁾, it seems that the solvent action of solvents is influenced by some physical factors such as polarity, dielectric constant, dipole moment and the degree of association in the molecules of the solvents. However, it may be difficult to interpret even the solvent action of solvents as physical action by only above factors. Furthermore, the carrier efficiency

of solvents is such a complex biological action that it may be difficult to interpret by the above factors.

As Hurst¹¹⁾ pointed out, a mixture of apolar and polar solvents was effective in increasing the permeability of Calliphorid larvae. We attempted to consider the correlation between the efficiency and the values of the above factors referred to by Kuwata¹⁰⁾. The results concerning the correlation are given in Tables 10 to 13. The same factors governing solubility affect the carrier efficiency of solvents but a clear relationship was not indicated. Therefore, further investigation is necessary in order to determine the limiting factors governing carrier efficiency.

Relationship between carrier efficiency and the molecular structure of primary alcohols

It is well known that the biological activity in a series of alcohols tends to increase with an increase in carbon atoms. Thus it is interpreted

Table 10. The relationship between carrier efficiency and the association degree of molecule of solvents against the oriental garden cricket and the housefly (3 replicate trials).

Solvent tested	The association degree of molecule of solvent	Log. values transformed the time of paralysis to the cricket		The steady mortality of the housefly	
		Female	Male	1%	2%
Nitrobenzene	0.93	1.4771	1.9395	42.0	45.1
Benzene	1.01	2.0719	0.0000	29.6	37.0
Aniline	1.05	1.6812	0.6021		
Acetone	1.26	2.5378	1.7924	20.0	38.7
<i>n</i> -Butyl alcohol	1.94	2.4742	2.2529	34.0	56.0
Ethyl alcohol	2.74	2.9191	2.4065	28.5	35.0
Ethylene glycol	2.92	2.1703	2.3766	48.8	46.0

Table 11. The relationship between carrier efficiency and the dielectric constant of solvents against the oriental garden cricket and the housefly (3 replicated trials).

Solvent tested	The dielectric constant of solvent	Log. values transformed the time of paralysis to the cricket		The steady mortality of the housefly	
		Female	Male	1%	2%
<i>n</i> -Hexane	1.85	1.4314	1.0414		
Dioxane	2.23	1.2553	0.8451	30.8	34.7
Carbon tetrachloride	2.24	2.2833	2.6191	36.0	40.3
Benzene	2.28	2.0719	0.0000	29.6	37.0
Toluene	2.39	1.2641	1.2788	29.4	34.5
Xylene	2.58	1.5911	1.6335	31.6	43.7
Trichloroethylene	3.40	1.2553	1.6812	40.3	51.9
<i>iso</i> -Amyl acetate	4.81	1.2553	0.6990	42.5	67.1
Butyl acetate	5.00	1.7993	2.3945	32.4	37.0
Chloroform	5.05	1.7782	1.7559	47.2	67.2
Monochlorobenzene	5.90	1.5185	1.9823	20.2	36.0
Ethyl acetate	6.12	2.4082	2.2967	37.4	32.7
Aniline	7.00	1.6812	0.6021		
Tri-cresol	10.10	2.7118	1.6232	34.3	61.9
<i>iso</i> -Butyl alcohol	18.90	2.1732	1.4150	46.0	73.0
<i>n</i> -Butyl alcohol	19.20	2.4742	2.2529	34.0	56.0
Acetone	21.40	2.5378	1.7924	20.0	38.7
Ethyl alcohol	25.80	1.9191	2.4065	28.5	35.0
Methyl alcohol	33.20	1.4914	1.5185	32.0	33.0
Nitrobenzene	35.70	1.4771	1.9395	42.0	45.1

that the biological activity results from the increase or the decrease on the factors such as volatility, viscosity, lipid solubility or surface activity. Hurst^{13,16} examined the effect of a homologous series of normal primary alcohols and fatty acids on blowfly larvae. He found that the molecular activity was roughly proportional to chain length. In injection tests, activity increased as the chain length of carbon atoms in the

alcohols increased from C₁ to C₅. In immersion tests, activity increased from C₁ to C₅ and thereafter from C₅ to C₈, a corresponding decrease occurred.

We attempted to consider the effect of primary alcohols tested against the housefly after a 2% solvent-dust application. The results are given in Figure 14. It is shown that the primary alcohol series shows an increase in the steady mortality

Table 12. The relationship between carrier efficiency and the dipole moment of solvents against the oriental garden cricket and the housefly (3 replicated trials).

Solvent tested	The dipole moment of solvent	Log. values transformed the time of paralysis to the cricket		The steady mortality of the housefly	
		Female	Male	1%	3%
Benzene	0.00	2.0719	0.0000	29.6	37.0
Carbon tetrachloride	0.00	2.2833	2.6191	36.0	40.3
<i>n</i> -Hexane	0.00	1.4314	1.0414		
Xylene	0.34	1.5911	1.6335	31.6	43.7
Toluene	0.40	1.2041	1.2788	29.4	34.5
Dioxane	0.45	1.2553	0.8451	30.8	34.7
Trichloroethylene	0.50	1.2553	1.6812	40.3	51.9
Chloroform	1.18	1.7782	1.7559	47.2	67.2
Aniline	1.55	1.6812	0.6021		
Monochlorobenzene	1.57	1.5185	1.9823	20.2	36.0
<i>n</i> -Butyl alcohol	1.66	2.4742	2.2529	34.0	56.0
Methyl alcohol	1.664	1.4914	1.5185	32.0	33.0
Ethyl alcohol	1.696	1.9191	2.4065	28.5	35.0
<i>n</i> -Propyl alcohol	1.699	1.6812	2.0899	38.8	46.0
<i>iso</i> -Butyl alcohol	1.790	2.1732	1.4150	44.0	73.0
Ethyl acetate	1.81	2.4082	2.2967	37.4	32.7
<i>iso</i> -Amyl acetate	1.82	1.2553	0.6990	42.6	67.1
<i>iso</i> -Amyl alcohol	1.82	2.0607	1.4914	23.2	36.0
Butyl acetate	1.84	1.7993	2.3945	32.4	37.0
Acetone	2.74	2.5378	1.7924	20.0	38.7
Nitrobenzene	3.95	1.4771	1.9395	42.0	45.1

Table 13. The relationship between carrier efficiency and the orientation polarization of solvents against the oriental garden cricket and the housefly (3 replicated trials).

Solvent tested	The polarity of solvent	Log. values transformed the time of paralysis to the cricket		The steady mortality of the housefly	
		Female	Male	1%	2%
Benzene	0.0	2.0719	0.0000	29.6	37.0
Carbon tetrachloride	0.0	2.2833	2.6191	36.0	40.3
<i>n</i> -Hexane	0.0	1.4314	1.0414		
Toluene	3.3	1.2041	1.2788	29.4	34.5
Chloroform	22.9	1.7782	1.7559	47.2	67.2
Aniline	47.5	1.6812	0.6021		
Methyl alcohol	58.6	1.4914	1.5185	32.0	33.0
Ethyl alcohol	60.1	1.9191	2.4065	28.5	35.0
Monochlorobenzene	60.3	1.5185	1.9823	20.2	36.0
Ethyl acetate	71.7	2.4082	2.2967	37.4	32.7
Acetone	163.0	2.5378	1.7924	20.0	38.7
Nitrobenzene	366.0	1.4771	1.9395	42.0	45.1

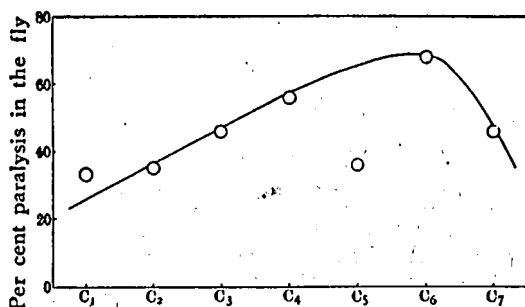


Fig. 14. Graph showing the effect of the series of primary alcohols on lindane dust. Lindane dust contains 2% of alcohols. The insect used: the adult of the housefly.

from chain length C₁ to C₆ and thereafter from C₆ to C₇, a corresponding decrease.

Joint toxic action between insecticidal solvents and lindane applied as a dust formulation

Sakai²⁵⁾ studied the joint toxic action between contact insecticides. In the present investigation, we attempted to examine several problems unanswered in the previous papers. The important point of the present paper is to determine whether the interactions between the solvents and lindane are due to independent joint action or the predominant activity of lindane. The present experiment could not determine whether or not there was any similar joint action between solvents and lindane.

We examined to calculate the predominant action of lindane alone instead of the similar joint action as follows.

The ratio of the predominant action of lindane

$$= \frac{\text{The mortality due to the mixture of lindane alone}}{\text{The mortality due to lindane}}$$

The above ratio was calculated to the mortality after 3, 5, and 18 hours, respectively.

We also examined to calculate the value of the independent action as follows: As proposed by Bliss²⁶⁾, and Plackett & Hewlett²⁷⁾, the expected mortality to the independent joint action of mixture is given by the following equations.

$$P = P_1 + P_2(1 - P_1)(1 - r) \quad (4)$$

where P is the expected mortality for the mixture of two constituents, P_1 , P_2 , the mortality

for two constituents respectively when used separately, r , the degree of association (the degree of correlation).

When the mixture occurs a complete positive correlation, if $P_1 > P_2$, thus if $r = 1$,

$$P = P_1 \quad (5)$$

When the mixture occurs a complete negative correlation, thus $r = -1$,

$$P = P_1 + P_2 \quad (6)$$

When the mixture occurs no correlation, thus $r = 0$,

$$P = P_1 + P_2(1 - P_1) \quad (7)$$

If the mixture consists of three or more constituents when the independent joint action indicates no correlation,

$$P = 1 - (1 - P_1)(1 - P_2)(1 - P_3) \dots (1 - P_n) \quad (8)$$

The present authors utilized equations (5), (6), and (8). When the equations (5) and (6) were used, the mixture regard to consist of the lindane and the solvent plus dust carrier.

Independent joint action-ratios were calculated by the following equation: Independent joint action-ratio

$$= \frac{\text{The observed mortality at the elapsed time in hours, respectively}}{\text{The expected mortality of independent joint action at the elapsed time in hours, respectively.}}$$

The values calculated for the expected independent joint action and for the predominant action to lindane were indicated in Table 14.

In conclusion it may be stated that the joint action between lindane and the solvents was indicated by a slight increase to a slight decrease of the carrier efficiency.

As shown in Table 14, in the case of the independent joint action-ratios applied with 2% dust, the higher joint action, in descending order of effectiveness, were indicated at the time of 18 hours as follows: 1) *iso*-butyl alcohol 2) cyclo-hexanol 3) methyl *iso*-butyl ketone 4) chloroform 5) tri-cresol 6) ethylene dichloride 7) *iso*-amyl acetate 8) trichloroethylene 9) methylethyl ketone 10) nitrobenzene.

In the case of the predominant action-ratios of lindane applied with 2% dust, the higher ratio,

Table 14. The theoretical values of the expected independent joint action between lindane and solvents and the calculated values of the predominant action of the mixture of lindane plus solvents to lindane alone against the housefly.

Code no. of solvent tested. Solvent content, % in w/w	The theoretical values of the expected independent joint action*									The calculated values of the predominant action to lindane		
	No correlation**			Independent joint action-ratio			Complete negative correlation					
	3 hr.	5 hr.	18hr.	3 hr.	5 hr.	18 hr.	3 hr.	5 hr.	18 hr.	3 hr.	5 hr.	18 hr.
k-1 1%	33.9	35.2	38.0	0.545	0.486	0.599	33.2	35.1	37.3	0.558	0.516	0.688
	2% 34.3	34.4	36.8	0.855	1.125	1.175	33.7	33.9	35.4	0.884	1.167	1.303
k-2 1%	33.7	34.2	35.6	0.895	1.038	0.995	34.4	33.6	33.5	0.938	1.071	1.068
	2% 34.7	34.7	37.7	1.042	1.298	1.169	34.3	34.3	36.7	1.089	1.357	1.327
k-3 1%	34.7	34.2	35.7	0.610	0.705	0.783	34.4	33.6	33.6	0.639	0.727	0.841
	2% 34.7	34.0	37.2	0.743	0.898	0.922	34.4	33.2	36.0	0.778	0.920	1.034
k-4 1%	34.5	34.0	37.9	1.416	0.990	0.951	33.2	33.2	37.0	1.472	1.074	1.086
	2% 36.2	36.3	38.4	1.376	1.228	1.097	33.2	36.7	37.8	1.502	1.342	1.270
k-5 1%	33.9	35.1	37.1	0.631	0.707	0.669	34.4	34.9	35.8	0.654	0.748	0.748
	2% 33.9	34.9	38.0	0.731	0.802	0.742	34.7	34.7	36.3	0.748	0.844	0.850
k-6 1%	34.7	34.3	35.7	0.818	0.981	1.011	34.4	33.8	33.7	0.856	1.061	1.089
	2% 34.9	34.9	36.4	1.111	1.154	1.166	34.7	34.7	34.7	1.170	1.285	1.279
k-7 1%	34.7	34.2	36.0	0.823	0.822	0.780	34.4	33.6	34.2	0.682	0.847	0.847
	2% 35.8	34.3	37.4	1.048	1.188	1.091	36.0	34.8	32.3	1.131	1.230	1.230
k-8 1%	34.7	35.1	37.5	1.276	1.252	1.117	34.4	34.9	36.5	1.336	1.324	1.114
	2% 36.9	37.4	49.4	1.671	1.741	1.394	36.9	38.5	54.9	1.834	1.966	2.078
k-9 1%	36.4	37.4	38.1	0.511	0.379	0.373	36.9	38.5	37.4	0.259	0.428	0.428
	2% 35.5	36.6	38.9	1.323	1.755	1.806	35.5	36.9	38.5	1.414	1.936	2.117
p-1 1%	33.9	34.4	36.6	0.772	0.869	0.849	33.2	33.9	35.1	0.790	0.902	0.938
	2% 34.4	34.5	37.9	0.808	0.778	0.888	33.9	34.1	37.0	0.838	0.808	1.013
p-2 1%	34.5	34.4	37.1	0.721	0.828	0.769	34.1	33.9	35.8	0.751	0.859	0.859
	2% 35.4	35.5	36.9	0.818	1.001	0.949	35.4	35.5	35.5	0.905	1.071	1.055
p-3 1%	34.5	34.5	40.6	1.126	1.131	0.969	34.0	34.0	34.0	1.170	1.176	1.185
	2% 35.4	35.4	38.2	1.164	1.283	1.227	35.4	35.4	37.6	1.242	1.369	1.414
p-4 1%	35.2	35.7	36.7	0.938	0.963	0.924	35.1	35.9	35.2	0.995	1.037	1.022
	2% 34.8	34.4	37.0	0.704	0.884	0.874	34.5	33.9	35.6	0.739	0.917	0.974
p-5 1%	35.1	35.1	36.4	0.958	0.961	0.972	34.9	34.9	34.8	1.013	1.016	1.068
	2% 36.5	38.2	40.2	1.476	1.467	1.382	37.1	40.0	40.7	1.625	1.692	1.677
p-6 1%	34.6	34.6	36.0	1.366	1.286	1.393	34.2	34.2	34.2	1.426	1.342	1.514
	2% 35.9	37.2	38.6	1.726	1.939	1.940	36.1	38.1	38.1	1.867	2.174	2.256
p-7 1%	34.4	35.3	37.1	0.695	0.633	0.606	33.9	35.2	35.9	0.721	0.672	0.672
	2% 35.0	36.7	39.3	0.957	0.933	0.917	36.2	37.3	37.2	1.010	1.032	1.086
p-8 1%	34.4	35.0	36.4	0.933	0.847	0.820	33.9	34.7	34.7	0.968	0.893	0.899
	2% 34.3	34.5	36.5	1.191	1.295	1.288	33.8	34.1	34.9	1.233	1.348	1.417
p-9 1%	34.5	35.6	37.0	1.427	1.144	1.192	34.0	35.7	35.7	1.484	1.227	1.330
	2% 36.2	37.0	39.2	1.829	1.852	1.844	36.6	37.9	39.1	1.996	2.069	2.180
h-1 1%	35.8	36.4	38.6	1.302	1.369	1.209	36.0	36.9	38.2	1.405	1.502	1.408
	2% 37.6	39.0	42.1	1.749	1.735	1.608	38.7	40.8	43.6	1.981	2.039	2.042
h-2 1%	34.5	34.5	36.0	0.933	1.031	1.001	34.1	34.1	34.1	0.971	1.034	1.086
	2% 33.3	37.0	40.0	1.194	1.101	1.003	35.2	37.9	40.0	1.197	1.230	1.233
h-3 1%	34.8	34.8	36.8	1.198	1.204	1.110	34.5	34.5	35.3	1.258	1.264	1.230
	2% 37.0	37.0	39.1	1.442	1.528	1.571	37.8	37.8	39.0	1.610	1.704	1.855
h-4 1%	34.6	35.5	39.7	1.099	1.053	1.020	34.2	35.6	39.9	1.196	1.128	1.221
	2% 38.2	38.2	41.6	1.292	1.253	1.414	39.7	39.7	42.8	1.490	1.445	1.773
h-5 1%	37.0	34.4	35.8	0.738	0.885	0.925	37.8	33.9	33.8	0.823	0.914	0.998
	2% 37.6	41.9	40.8	1.129	0.992	1.119	38.8	45.3	41.5	1.282	1.255	1.375
s-1 1%	34.8	33.0	37.8	1.184	1.150	0.952	34.5	33.2	33.8	1.242	1.146	1.086
	2% 37.4	39.8	45.2	0.771	0.718	0.750	38.4	42.1	48.4	0.869	0.862	1.022

防 虫 科 学 第 22 卷—I

s-2	1%	35.2	35.1	37.3	0.432	0.504	0.478	35.1	35.0	26.1	0.458	0.534	0.537
	2%	36.8	36.8	93.6	0.957	1.096	0.907	37.5	37.5	39.7	1.083	1.215	1.083
s-3	1%	34.5	34.4	35.9	0.890	0.945	0.863	34.1	33.9	34.0	0.926	0.980	0.935
	2%	34.3	36.2	36.5	1.034	1.000	1.024	33.8	26.6	34.9	1.071	1.092	1.128
s-4	1%	38.1	45.1	37.5	1.118	0.945	1.120	29.5	34.9	26.5	1.285	1.285	1.279
	2%	40.7	41.5	44.4	1.524	1.615	1.510	43.4	44.7	47.2	1.882	2.024	2.024
g-1	1%	35.5	34.9	36.3	0.525	0.571	0.570	35.5	34.6	34.6	0.561	0.600	0.624
	2%	34.7	34.4	35.8	1.037	1.166	1.119	34.4	33.9	33.9	1.086	1.209	1.209
g-2	1%	41.9	47.3	37.1	0.301	0.287	0.361	44.2	38.3	35.9	0.380	0.410	0.404
	2%	38.3	37.0	36.9	0.608	0.726	0.751	40.2	39.8	35.4	0.703	0.820	0.835
g-3	1%	34.0	34.0	36.1	0.595	0.657	0.983	33.2	33.2	34.3	0.609	0.672	1.071
	2%	33.9	33.9	37.5	1.096	1.235	1.170	33.2	33.2	36.5	1.122	1.264	1.324
g-4	1%	35.3	35.3	36.7	1.399	1.384	1.246	35.2	35.2	35.2	1.487	1.472	1.378
	2%	37.6	38.2	39.5	1.182	1.205	1.270	38.7	39.6	39.6	1.339	1.387	1.514
g-5	1%	34.5	34.5	36.2	0.464	0.628	0.840	34.0	34.1	34.4	0.483	0.654	0.917
	2%	35.3	34.9	42.2	0.618	0.598	0.647	35.2	34.7	43.7	0.657	0.630	0.823
g-6	1%	34.6	34.6	37.8	0.720	1.017	1.091	34.2	34.2	36.9	0.751	1.062	1.242
	2%	35.1	34.8	38.0	0.359	0.333	0.463	34.9	34.5	37.3	0.380	0.350	0.531
b-1	1%	34.7	34.7	37.9	0.775	0.780	0.801	34.4	34.4	37.0	0.814	0.817	0.914
	2%	34.6	35.9	39.8	0.725	0.919	0.955	34.2	36.2	40.0	0.757	0.995	1.146
b-2	1%	34.7	36.4	37.3	0.481	0.492	0.597	34.4	37.2	36.2	0.504	0.540	0.672
	2%	35.1	36.6	39.4	1.082	0.929	0.914	35.0	34.2	39.4	1.146	1.025	1.086
b-3	1%	35.3	35.9	36.6	1.141	1.203	1.177	35.3	36.2	35.1	1.215	1.303	1.300
	2%	35.3	38.0	37.9	1.277	1.222	1.332	35.3	39.4	37.1	1.360	1.402	1.523
b-4	1%	34.6	34.6	37.3	0.829	0.942	0.876	34.2	36.7	36.2	0.865	0.983	0.986
	2%	35.5	36.3	37.8	0.985	0.960	0.985	35.6	36.7	37.9	1.055	1.049	1.122
b-5	1%	34.7	35.1	37.8	0.884	0.808	0.805	34.4	35.0	36.9	0.926	0.856	0.917
	2%	37.0	38.2	41.5	0.664	0.824	0.828	37.9	39.7	32.7	0.742	0.950	1.037
b-6	1%	34.5	34.5	37.4	0.909	0.967	0.896	34.1	34.1	36.3	0.947	1.007	1.010
	2%	34.8	34.7	37.7	0.549	0.857	1.009	34.3	34.3	36.7	0.576	0.896	1.145
b-7	1%	35.5	37.2	39.0	0.338	0.961	0.800	35.5	38.2	38.8	0.362	1.086	0.941
	2%	35.7	37.7	41.9	1.310	1.119	1.018	35.8	38.9	43.2	1.408	1.273	1.285
b-8	1%	35.5	35.5	36.9	0.846	0.905	0.968	35.5	35.5	35.5	0.905	0.968	1.077
	2%	38.1	35.5	39.2	1.162	1.720	1.589	39.5	35.6	39.1	1.870	1.843	1.879
b-9	1%	35.8	37.0	42.2	0.704	0.837	0.682	36.0	37.9	43.8	0.769	0.935	0.869
	2%	37.9	38.6	43.0	0.847	0.898	0.818	39.2	40.3	45.0	0.968	1.046	1.062
b-10	1%	36.2	34.6	35.9	0.915	1.038	1.114	36.6	34.2	34.0	0.998	1.086	1.098
	2%	39.0	36.9	39.1	1.030	1.221	1.177	40.9	37.6	38.9	1.212	1.357	1.387
b-11	1%	34.3	34.3	36.4	1.727	1.141	0.990	33.7	33.7	34.7	1.785	1.179	1.086
	2%	50.2	48.9	50.1	0.534	0.564	0.719	58.6	55.9	55.9	0.808	0.833	1.086
e-1	1%	34.7	35.1	36.8	0.855	0.881	0.850	34.4	34.9	35.4	0.896	0.932	0.944
	2%	34.7	35.1	35.9	0.942	0.927	0.972	34.4	34.9	34.0	0.986	0.990	1.052

* The complete positive correlation between lindane and solvents was equal with the mortality of lindane alone.

** The theoretical value when the mixture occurs no correlation, was calculated from the value of lindane, solvent and dust carrier, separately.

in descending order of effectiveness, were 1) *iso*-butyl alcohol 2) cyclohexanol 3) chloroform 4) methylethyl ketone 5) tri-cresol 6) ethylene dichloride 7) *n*-butyl alcohol 8) trichloroethylene 9) alpha-pinene 10) *n*-propyl alcohol, acetophenone and ethylene glycol.

Discussion

In the authors' opinion, the carrier efficiency

of organic solvents may be an important factor in insecticidal action from both a theoretical and practical aspects.

It is interesting that the carrier efficiency of solvents is probably an essential factor in the demonstration of joint toxic action of insecticides.

Considering the above fact, the present authors attempted to study the carrier action and the joint toxic action of solvents.

The present investigation, however, did not result in a determination of a clear relationship to the carrier efficiency of solvents probably because of the poor design of the experiments and from many unknown factors governing the carrier efficiency.

Although the present investigation was examined by the application of dust formulation against the housefly in order to exclude the problems caused by the use of water, further investigation should be made to examine these problems by such means as topical application to individual insects and the use of the immersion test using liquid formulation diluted by water.

Further investigation should be attempted to analyse the water-solvent-detergent systems of the active insecticides.

Although the present investigation avoided the toxic action of lindane vapour alone as a respiratory poison, the next study should consider the interaction between cuticular penetration and tracheal penetration of vapours.

As pointed out by Hurst¹¹⁾, the combination of solvents, such as the mixture of apolar and polar solvents differs from a simple solvent by solvation. Therefore, the solvation between solvents should be examined from the viewpoint of insect toxicology.

Summary

The present investigation attempted to demonstrate the joint toxic action between organic solvents and lindane and to examine the effects of fifty eight solvents on the penetration of lindane powders.

The insects used were the adult of the common housefly, *Musca domestica vicina* Macq., and the adult of the oriental garden cricket, *Gryllus mitratus* de Saussure.

The insecticide adopted was lindane. Fifty eight insecticidal solvents were tested. The dust carrier mixture consisting of 6-parts clay and 4-parts talc (300 mesh) was applied to the dust formulations.

Air-vacuum dust apparatus was utilized for the dusting of the housefly. In the case of the cricket test, the tarsus of the cricket's hind leg

was immersed into the test solution for 30 seconds. The per cent paralysis in the insects were observed.

In the experiments on the relation between time and per cent paralysis in the housefly for the lindane dusts added to the solvents, the higher carrier efficiencies of the solvents adopted, in descending order of effectiveness, were expressed as follows: 1) chloroform 2) *iso*-butyl alcohol 3) ethylene glycol 4) methylethyl ketone 5) ethylene dichloride 6) cyclohexanol 7) *iso*-amyl acetate 8) nitrobenzene 9) *n*-propyl alcohol 10) cyclohexanone.

In the case of the oriental garden cricket, 1) *iso*-amyl acetate 2) dioxane 3) toluene 4) *o*-chlorotoluene 5) cyclohexanol 6) methyl *iso*-butyl ketone 7) Velsicol AR-60 8) *n*-hexane 9) Penn Drake oils 10) trichloroethylene.

Combining the values obtained for the cricket with that of the housefly resulted in the carrier efficiencies of the solvents, the following descending order of effectiveness was obtained: 1) *iso*-amyl acetate 2) cyclohexanol 3) tri-chloroethylene 4) methyl ethyl ketone 5) cyclohexanone and methyl *iso*-butyl ketone 7) *iso*-butyl alcohol, *o*-chlorotoluene and *n*-hexane 10) dioxane.

The aromatic hydrocarbons showed the efficiency. Aliphatic ketones and halogenated hydrocarbons were moderately efficient. Generally speaking, glycols and its derivatives, the esters of acetic acid and aliphatic alcohols had low efficiencies.

The present investigation also attempted to determine a tendency to a general relationship between carrier efficiency and boiling point of solvent. At a glance, the line to the cricket tested were generally indicated the line curved like a bowl and the lines to the housefly tested were generally indicated the line curved like a mountain with the exception of some solvent-groups.

Although a clear relationship to the solubility to lindane, beeswax, and the housefly's lipoids, was not apparent, the higher solubility to beeswax and the housefly's lipoids of solvents indicated the comparatively higher carrier efficiency by trichloroethylene, cyclohexanol, cyclohexanone, *o*-chlorotoluene.

The relationship between carrier efficiency and the surface tension of solvents saturated with lindane against the crickets was more obvious than the relationship against the housefly. However, there were many unfavourable results with both insects. There was a comparatively clear relationship in the female cricket. The speed of paralysis gradually reduced with an increase of the surface tension for the experiments of aliphatic primary alcohols, aliphatic ketones and aromatic hydrocarbons.

In the experiments for the relationship to the viscosity of solvents at a temperature of 20°, the results on the male cricket and the housefly applied with 1% dust gave the fairly clear relationship to the viscosity.

The carrier efficiency of solvent was difficult to interpret by some physical factors such as polarity, dielectric constant, dipole moment and the association degree of molecules of the solvents. Therefore, further investigation is necessary in order to determine the limiting factors governing carrier efficiency.

The primary alcohol series tested show an increase in the steady mortality from chain length C₁ to C₆ and thereafter from C₆ to C₇, a corresponding decrease.

In conclusion, it may be stated that the joint action between lindane and the solvents tested was indicated by a slight increase to a slight decrease of the carrier efficiency. In the case of the independent joint action-ratios applied with 2% dust, the higher joint action, in descending order of effectiveness, were indicated at the time of 18 hours as follows : 1) *iso*-butyl alcohol 2) cyclo-hexanol 3) methyl *iso*-butyl ketone 4) chloroform 5) tri-cresol 6) ethylene dichloride 7) *iso*-amyl acetate 8) trichloroethylene 9) methyl-ethyl ketone 10) nitrobenzene. In the case of the predominant action-ratios to lindane applied with 2% dust, the higher ratio, in descending order of effectiveness, were as follows : 1) *iso*-butyl alcohol 2) cyclohexanol 3) chloroform 4) methyl-ethyl ketone 5) tri-cresol 6) ethylene dichloride 7) *n*-butyl alcohol 8) trichloroethylene 9) alpha-pinene 10) *n*-propyl alcohol, acetophenone and ethylene glycol.

The present investigation did not obtained a clear relationship of the carrier efficiency of solvents probably because of a poor experimental design and from many actions of complicating factors governing carrier efficiency. Therefore, further investigations are required to solve these problems.

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Analytical Observations on the Lethal Doses of DDT and BHC to Mosquito Larvae. Studies on the Action of Insecticides. II. Shigeo HAYASHI and Takeshi SUZUKI (Department of Parasitology, Institute for Infectious Diseases, University of Tokyo, Tokyo). Received Nov. 11, 1956. *Botyu-Kagaku*, **22**, 138-144, 1957, (with English résumé, 144).

21. 蚊幼虫に対する DDT 及び BHC の致死量についての解析 殺虫作用に関する研究 第2報 林 滋生・鈴木 猛(東京大学 伝染病研究所 寄生虫研究部) 31. 11. 11 受理

謹んで春川忠吉博士の古稀を祝賀し奉る。

蚊幼虫に殺虫剤を食毒として与えた場合、各個体についてそれぞれの致死量を簡便に測定出来るようにした。この致死量と薬剤濃度の間の関係を追求したところ、薬剤の作用という刺激とそれに対する反応との間に、Weber-Fechner の法則が適合する場合には、各個体の感受性は、薬剤濃度そのものではなく、その対数に対して正規分布をすることが理論的に帰結された。なお、DDT, BHC, リンデンの実験例をあげ、致死量にあらわれた薬剤の特性を比較考察した。

殺虫剤の効力試験にあたって、その結果の統計的処理には、現在 Bliss^{1,2)} の方法が広く用いられている。これは、Trevan¹⁰⁾, Gaddum⁹⁾ らの系統をひく考え方に立脚し、我が国では、大沢・長沢⁹⁾ の綜説によつて紹介されて以来、長沢はじめ、多くの研究者によつてとり上げられている。Bliss の方法は、しばしば指摘されているごとく、その根本に、作用曲線感受性の個体差分布の累積曲線と解釈すること、また濃度を適当に変換するとこの分布が正規に近くなることを仮定している。そして、この後者の仮定については、若干の検討が加えられてはいるが(たとえば河野⁶⁾)、多くの場合に、単に経験的な事実から、濃度を対数に変換しているにすぎない。

しかし、対数変換しても正規に近づかない場合、すなわち、プロビットを用いていわゆる1次変換をしても直線にならない場合がしばしばある。これがふたつあるいはそれ以上の直線の連続によつて表現され、作用機序の異なるものの合成として理解される場合もあるが、またそのように考えられない場合も多い。

本稿に報告する実験は、経験的に知られた対数変換

が、何故正規になるか*。またひいては、どういう場合に対数変換が不適当か、ということを解明したい意図から出発している。本実験の基本的な部分の一部は、さきに筆者の一人林によつて予報されたが⁴⁾、更にこの実験方法を応用発展させることによつて、興味ある知見を得つゝあるので、逐次報告してゆく予定である。

この研究に御指導を頂いた伝染病研究所寄生虫研究部佐々学助教授、実験を担当して頂いた木村マリ、佐藤金作、松永秀子諸氏に厚く感謝の意を表する。

実験材料及び方法

通常の殺虫試験では、供試昆虫の薬剤に対する感受性の個体差分布を、直接求めることが出来ない(時間・致死率などの試験では、ある程度可能ではあるが)。このため、濃度・致死率の実験においては、いくつかの段階の濃度における致死率をしらべ、いわゆる作用曲線を求めて、これを感受性分布の累積曲線として分析をすすめるわけである。

* 第1報以後、金光・長谷川⁵⁾ が別種の説明を試みた報告があらわれた。